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PROCEEDINGS OF INTERNATIONAL CONFERENCE ON FLUIDIZED-BED COMBUSTION (2nd)

Environmental Protection Agency
Research Triangle Park, North Carolina

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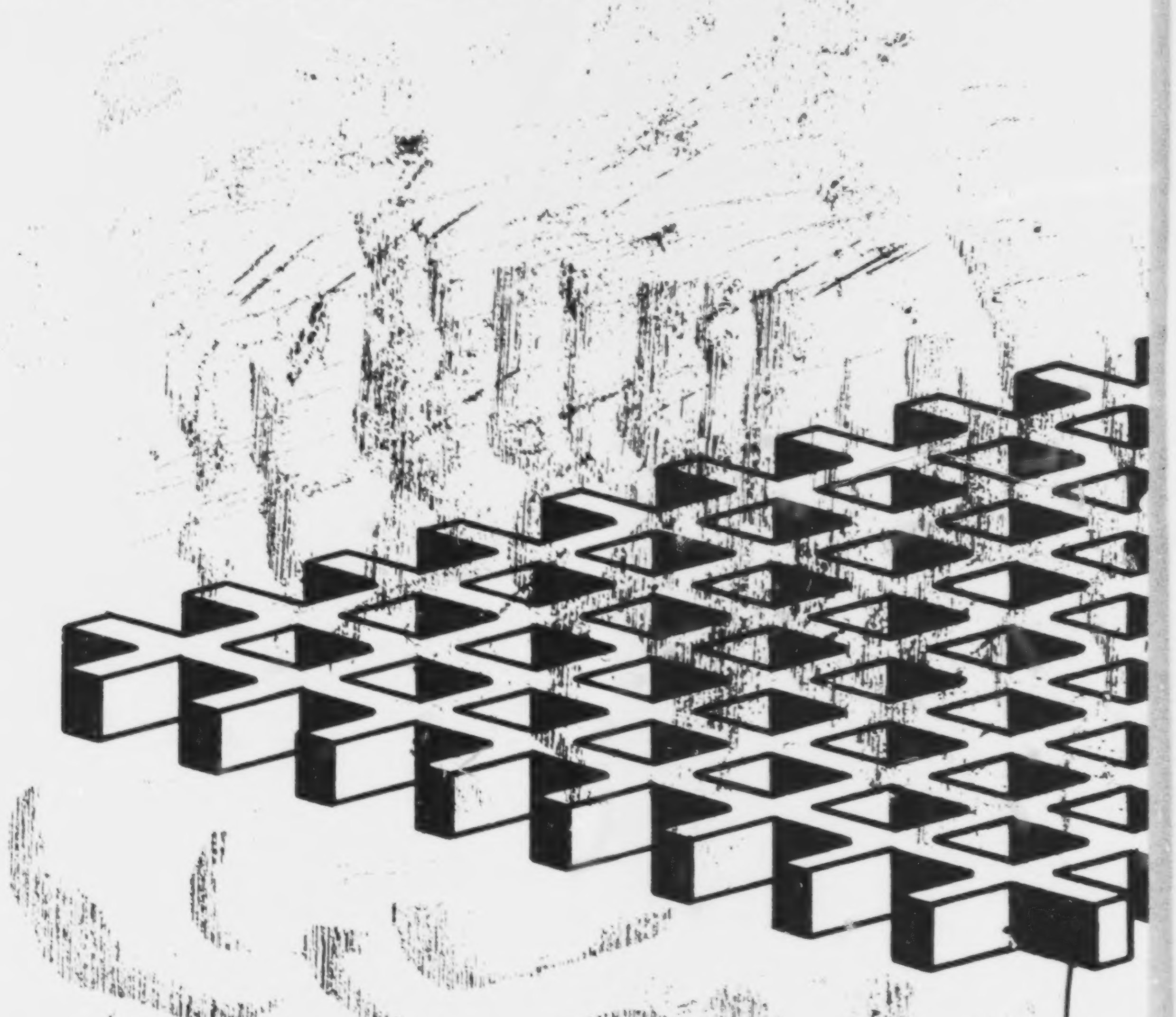
OF SECOND INTERNATIONAL CONFERENCE ON FLUIDIZED-BED COMBUSTION



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October 4, 1970

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U.S. ENVIRONMENTAL PROTECTION AGENCY
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PREFACE

The Second International Conference on Fluidized-Bed Combustion was held October 4-7, 1970, at Hueston Woods Lodge, RFD No. 1, College Corner, Ohio, under the sponsorship of the U.S. Department of Health, Education, and Welfare's National Air Pollution Control Administration (NAPCA). Subsequent to the Conference, NAPCA's functions were transferred: first, to the Air Pollution Control Office (APCO) of the newly created Environmental Protection Agency (EPA); later to EPA's Office of Air Programs (OAP). All references to NAPCA in these proceedings, therefore, now refer to OAP.

The Conference, under the general- and vice-chairmanship of Messrs. P.P. Turner and D.B. Henschel, respectively, was open to early arrivers Sunday, October 4.

The Conference proper, consisting of six sessions, got underway Monday morning with the official welcome extended by OAP's Sheldon Meyers. Following Mr. Turner's introductory remarks, Prof. D.E. Elliott delivered the keynote address, "Exploiting Fluidised-Bed* Combustion."

The six-part Session I, chaired by Mr. A.A. Jonke, followed the theme, "Small-Scale Development of Fluidized-Bed Combustion." Session II, held Monday evening, also consisted of six presentations, this time on the general topic, "Control of Combustion Pollutants;" Mr. Alvin Skopp was chairman.

Mr. J.W. Eckerd was chairman of Session III, titled "Gasification to Desulfurize Coal, " which opened Tuesday's activities. This session consisted of seven presentations. "Conceptual Design and Economic Feasibility" was the theme of both Sessions IV and V. The former, chaired by Dr. D.H. Archer, consisted of five papers presented Tuesday evening: the latter, by Mr. T.C.L. Nicole, consisted of six papers presented Wednesday morning.

Session VI, the last of the Conference, was a Wednesday afternoon discussion, led by a panel of six, summarizing thoughts presented during the Conference and providing a final opportunity for comments from the floor. Chaired by Mr. R.P. Hangebrauck, the panel consisted of Prof. Elliott, Dr. Archer, Dr. C.Y. Wen, Mr. Shelton Ehrlich, and Mr. Henschel.

All papers presented during the Conference are included in these proceedings.

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*In these proceedings, papers retain local spellings, where applicable.

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Mr. P. P. Turner, NAPCA

KEYNOTE ADDRESS:

Professor D. E. Elliott, University of Aston, England

Introductory Remarks

1. THE NAPCA-DPCE PROGRAM TO CONTROL POLLUTION FROM STATIONARY SOURCES

P.P. TURNER

Division of Process Control Engineering
National Air Pollution Control Administration

By way of review and to lay the groundwork for this conference, I want to present a bit of background regarding our reason for being here. The "First International Conference on Fluidized Bed Combustion" was sponsored by the National Air Pollution Control Administration and was held here in November 1968. Apparently the Conference was successful because it was decided that a similar meeting would be beneficial after 2 years of additional research. That decision has brought us together today for a further exchange of information.

In his introductory remarks to the first Conference, Mr. Paul W. Spaite (then Chief of the Process Control Engineering Program) explained the growth of U.S. interest and effort in controlling air pollution, specifically controlling the oxides of sulfur and nitrogen, as well as particulate matter, from stationary sources.

It is generally agreed that fluidized-bed combustion (FBC) has considerable pollution control potential. The lower FBC operating temperature (1400-1900°F) may minimize the formation and emission of nitrogen oxides (NO_x), and is near optimum for the reaction of SO_2 with limestone. Particulates can be removed by cyclones, scrubbers, or precipitators.

Removal of SO_2 remains a very serious problem. Table 1 gives some idea of the magnitude of the problem. The figures shown for 1970 and beyond are projected from estimates made by coal and oil interests and by industry. The tabulation shows that power plants emit most of the SO_2 that is in the atmosphere.

The demand for electric power is increasing so rapidly that sulfur oxides (SO_x) emissions may increase even if the following techniques are developed and applied as soon as possible:

1. Construction of nuclear power plants.
2. Substitution of gas or low-sulfur fuel oil or coal, where available.
3. Use of cleaned low-sulfur-content coal.
4. Introduction of improved combustion methods.
5. Application of improved stack gas treatment and sulfur recovery processes.

Even with a national commitment to orderly, but urgent, application of new technology, the best that we can hope for through the year 2000 is a total SO_x emission rate from all utilities near the present level. The number of plants burning fossil fuels is increasing that much.

Nuclear power plants emit no SO_2; essentially the same can be said for plants using natural gas. The technology for removal of sulfur from fuel oil appears to be reasonably well in hand based on Esso Research in England. Further development of hydroelectric power will not be a major factor. Importing liquified petroleum gases will most likely increase to the limit of economic availability. However, these methods of producing energy will not be sufficient. The use of coal will steadily increase: it is expected to more than triple by the year 2000, before leveling off as large nuclear power stations replace those burning fossil fuels. About 65 percent of the SO_x discharged into the atmosphere from man-made sources comes from the combustion of coal. The average sulfur content of coal now burned is 2.7 percent: this average is expected to increase to 3.5 percent by the year 2000, aggravating the situation additionally.

Table 1. ESTIMATED POTENTIAL SULFUR DIOXIDE POLLUTION IN THE U.S., WITHOUT ABATEMENT[a]

Source	Annual Emission of Sulfur Dioxide, millions of tons				
	1967	1970	1980	1990	2000
Power plant operation (coal and oil)	15.0	20.0	41.1	62.0	94.5
Other combustion of coal	5.1	4.8	4.0	3.1	1.6
Combustion of petroleum products (excluding power plant oil)	2.8	3.4	3.9	4.3	5.1
Smelting of metallic ores	3.8	4.0	5.3	7.1	9.6
Petroleum refinery operation	2.1	2.4	4.0	6.5	10.5
Miscellaneous sources[b]	2.0	2.0	2.6	3.4	4.5
TOTAL	30.8	36.6	60.9	86.4	125.8

[a] February 1970 estimates by National Air Pollution Control Administration, excluding transportation.

[b] Includes coke processing, sulfuric acid plants, coal refuse banks, refuse incineration, and pulp and paper manufacturing.

NAPCA's Division of Process Control Engineering (DPCE) is working on many techniques for eliminating SO_2 emissions to the atmosphere. Without elaborating on the various techniques, Table 2 gives you some idea of the scope of our program. Note the heading "Fluidized-bed combustion (FBC)" under item 3.

DPCE first undertook work on FBC in FY 1968 at a funding level of $400,000; the current effort is 5 times greater. NAPCA has already spent about $2.25 million of an estimated $75 million that will be required to

Table 2. PROCESS DEVELOPMENT FOR ABATEMENT OF SO_2

1. General considerations of individual processes
2. Precombustion processes
   - a. Coal cleaning
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3. Combustion processes
   - a. Fluidized-bed combustion (FBC)
   - b. The Black, Sivalls, and Bryson
4. Limestone processes
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   - e. Formate scrubbing
   - f. Ammonia scrubbing
   - g. Westvaco char
   - h. Molten carbonate
   - i. Sodium bicarbonate adsorption
   - j. Modified Claus
   - k. Catalytic chamber
   - l. Ionics/Stone & Webster
   - m. Alkalized alumina
6. Scrubber development

demonstrate the best FBC system. NAPCA believes that the FBC concept is particularly well-suited to collaborative research so that substantial portions of the total R & D expense can be shared by a number of government and industry groups both in the U.S. and abroad.

NAPCA's interest in FBC because of its pollution control potential brings us to SO_2 formation and emission when coal and oil are burned. Limestone addition for SO_2 control should be especially effective because fluidized-bed combustors operate in the temperature range best suited for the limestone-SO_2 reaction (1400-1900°F); the fluidized-bed combustor also should provide good mixing and longer residence time for the reaction to take place.

The current NAPCA program on FBC consists of laboratory and small pilot-scale research studies. In addition to work abroad, NAPCA projects on the air pollution control aspects of FBC are underway both at Pope, Evans and Robbins and at the Argonne National Laboratory. NAPCA is also sponsoring new work on FBC combustion and pollution control at: Esso Research and Engineering Co., Linden, N.J.; The U.S. Bureau of Mines, Morgantown, W. Va.; Westinghouse Research and Development Center; Consolidation Coal Co.; Esso Petroleum Co., Ltd.; and the National Coal Board, England.

NAPCA's future program includes conceptual design studies as well as technical and cost feasibility studies of FBC systems. Since each NAPCA-DPCE contractor is represented at this meeting, I will say nothing more about their programs; rather, I will let them present the results of their own work.

In closing, just a word about the various sessions. I have asked each Session Chairman to control his session carefully. The last speaker in a given session deserves as much time as the first; however, we also wish for in-depth discussion. The reason for limiting the participants in this conference is to stimulate discussion, both formal and informal, much as is done at the Gordon conferences – so let us hear from each of you.

It is now my pleasure to introduce our keynote speaker who will address the conference on "Exploiting Fluidized-Bed Combustion." He will point out the potential of this combustion concept including history, status, and prospects. He is well qualified to set the tone for this conference. He was with the Central Electricity Generating Board for 9 years working in the area of FBC heat transfer and carbonization of coal. For the past 2 years he has been Professor of Mechanical Engineering at the University of Aston in Birmingham, England, where he has continued his research on FBC concepts. Gentlemen, I present to this conference Professor Douglas E. Elliott.

Keynote Address

2. EXPLOITING FLUIDISED-BED COMBUSTION

D.E. ELLIOTT

University of Aston, England

INTRODUCTION

Although the original aims of the British and American fluidised-bed combustion research programmes were different, both programmes have moved along similar lines. The British saw fluidisation as a means of reducing the capital costs of power stations; the Americans placed much more emphasis on the possibilities of sulphur removal. However, the new technology has so many interesting and potentially commercial facets that we can now look forward to a period in which more and more effort will be mounted by a greater number of firms and research organisations on diverging projects. It is therefore useful at this stage to take stock of our knowledge, pinpoint problems that it is imperative to solve, estimate the probable return from competing processes, and put in hand the next round of developments.

In terms of the S-curve of Figure 1, characterizing the development of any technology, we are just at the toe. Whether fluidised-bed combustion peters out (a), as M.H.D. seems to have done, progresses at a modest rate (b), or escalates at an ever-increasing rate (c) depends mainly upon:

1. Is the technology based on good principles and are the natural phenomena working with us? Unless this is so no amount of effort will produce a workable plant. In my opinion we are not in much danger on this account and most of our encouraging results can be explained in sound scientific terms. Only in a few areas do we find conflicting requirements; in most of these there are alternative ways around the problems.
2. The choice and design of the pilot-scale plants which should follow our large-scale rig experiments. It is in this area that we are most vulnerable. A bad decision at this stage could mar the progress of fluidised-bed combustion, possibly irreparably. We cannot expect to be allowed to fritter away millions of pounds on this work, as has been the case with nuclear power, even though several abortive attempts could probably be justified on a cost/benefit basis.
3. The amount of ingenuity we apply and whether we are prepared to take more than one step at a time. There is always a reluctance on the part of the designer to do more than one thing at a time; in this day and age, this could lead to being overtaken by events.

This Second International Conference is therefore taking place at what must be the most critical period in the history of fluidised-bed combustion and some indication of the probable outcome will be the progress that has been made in the last year. Are our problems still the same as they were a year ago? Have we solved any major snags? Have any new ideas come forth? Are the economics turning out as was forecast? In view of the large number of technical papers which we shall be discussing this week, it would be pointless for me to do more than a brief survey; I would like to air a number of uncertainties in the hope that they will arouse future exchanges. I also wish to outline some exciting fields for further research.

PRESENT STATE (PRIOR CONFERENCE)

Heat Transfer

We know enough about the theory and the practical realisation of high heat-transfer coefficients to be certain that early claims of reduced heat-transfer surfaces in fluidised systems are fully justified. Little more work is needed here except for special conditions imposed by some advanced designs.

Fouling and Corrosion

Encouraging experience with heat transfer surfaces in rig testing over accumulated times of several hundred hours supports the contentions that this is one of the most important aspects of fluid-bed combustion. Long duration trials of 1000 hours or longer will be needed before we can assess the benefits of reduced maintenance and the possibility of achieving higher steam conditions, but there is good reason for optimism, at least when burning coal. Because there is little relevant information, work should start on the fouling and corrosion in heavy oil burning systems.

Combustion

I anticipate that we shall be hearing that with medium velocities using 1/8-in. coal, good combustion efficiencies can be obtained using an acceptable recycle ratio of fines, at temperatures of 1400 to 1500°F. The problem is not quite as clear for velocities of 8 to 14 ft/sec favoured by plant manufacturers because they cut down containment costs and produce compact units. Using larger coal entails a much higher carryover; also, recycling to the primary bed may give unreasonably high loadings of the separaters. Thus attention is being drawn to secondary beds for burning out the elutriated fines. Having successfully burnt the residue from anthracite burning power stations, which contained only about 20 percent carbon, I was surprised to hear that burning the fly ash from fluidised-bed combustors proved difficult in early tests. I am looking forward to the discussion of this problem; I can hardly believe that it is a critical one.

Carryover

While on the subject of carryover, I feel that too little attention has been paid to what happens to the coal immediately after its injection into the bed. There is plenty of evidence to show that, under rapid heating conditions, the volatile emission is much higher than that shown by standard assays and that the swelling of the particles is increased. Consequently, there is a greater tendency for the particles to decrepitate on carbonising and the strength of the chars is lower; both lead to greater degradation of the particles by attrition in the bed. It is therefore useful to speculate if we could improve matters by some preconditioning of the coal or by making sure that we do not have too rapid heating.

Coal Feed

One of the two most serious problems still outstanding is coal distribution. I have seen tentative designs with feed points 10 feet apart. Do we really think this is practical? If not, how close must they be? From time to time, people talk about the rapid mixing in fluidised beds; because lateral mixing is relatively slow and to prevent massive local evolution of volatiles, we must have a good spreading of coal. I will mention possible solutions later.

Startup and Control

The second serious problem (the first was coal feed) arises because the heat extraction rate of submerged tubes only falls a little at low fluidising velocities and because combustion cannot take place effectively at temperatures much below 1300°F. Thus we cannot start a bed containing a full array of tubes unless we provide a burner of near-full rating beneath the bed. Similarly, we can only vary the output of such a bed over a very narrow range of output and resort to separate beds and startup beds without tubes.

Base Design

Although there may be room for improvement, base design is not a major problem. Operating experience with large rigs suggests that acceptable designs with low pressure drops have been developed.

Sulphur Removal

Sulphur removal experience on either side of the Atlantic seems to vary. With British coals in British rigs, adding limestone retains a substantial proportion of the sulphur. I understand that American results have been rather disappointing. Whether this is because American coal is awkward, because American limestone is unreactive, or because American rig conditions vary will be established by testing American coal in British apparatus. The fact that we have one set of encouraging data suggests that some way round this problem will be forthcoming.

The economics of the sulphur removal have yet to be disclosed. Can we afford to add limestone in the quantities required? Can we dispose of the resulting ash without polluting the land? Would it be better to go for a regeneration system? What can we do about the existing power stations? Can we do for coal what Esso is trying to do for heavy oil? Is it more economical to scrap existing boilers, replacing them with fluid-bed units? Many questions need to be answered.

Hydrocarbon Pollution

It is generally assumed that exhaust gases from fluidised beds will be very low in unburnt hydrocarbons. Experience supports this; but again, I return to the coal feed arrangements. Too much coal in any one area will almost certainly yield unburnt volatiles. On the credit side however, it is likely that the heavy clouds of smoke which accompany load changes in normal plants can be avoided in the new system. The fluidised bed has a high thermal inertia which should allow load changes to be met without violent changes in combustion conditions.

There is however another point which must be borne in mind. Will there be any very fine carbon produced which will evade the precipitators, and will the removal of sulphur from the gases reduce the efficiency of these units? It is natural to assume that, since we are starting with a larger size of coal, we must end with a larger size of dust which will be more readily caught. However, remember that it is not the mean size that matters: anything above 5 microns is easily captured. The submicron particles are what matter: they are more likely to be a function of the coal heating period and the attrition in the bed.

Oxides of Nitrogen

Because fluidised-bed combustion can take place at temperatures of 1500°F or below, then theoretically the yield of oxides of nitrogen should be very low; much lower than with adiabatic combustion where flame temperatures of 3000°F are common; and very much lower than is formed in petrol or diesel engines.

However, there have been conflicting reports of experimental data. Some of these suggest that the oxides of nitrogen are much above the theoretical equilibrium at the fluidised-bed temperature; others suggest that they are only marginally above this level. A possible explanation is that the particle temperature is several hundred degrees (F) higher than the bed temperature. Experiments, in which thermocouples have been placed in large (1/4-in.) particles, show this to be the case. This explanation is also confirmed by visual observation of fluidised combustors, especially those running with large coal at high velocities.

In this context, it is reasonable to expect that large-coal/high-velocity systems will be worse than smaller-coal/lower-velocity beds because:

1. There is a tendency to run high-velocity systems at a higher temperature to obtain acceptable combustion efficiency.
2. The scrubbing velocity of the air past the larger particles is higher; therefore, the reaction intensity will be higher.
3. The heat transfer from larger particles to their neighbouring large particles will be lower than when small particles are in the vicinity of other small particles.
4. The bed volume used in high velocity combustors is generally smaller than with low velocity units, hence the spatial density of the carbon bearing particles is higher and the chance of their coming together to create a temporary "hot spot" is greater.
5. Observation of fluidisation reveals that its quality is much smoother with small particles and, because lower bed depths can be used, the bubble size is smaller. Because bubbles contain few particles, temperatures therein can rise by the combustion either of volatiles or of the few particles in the bubbles.

Thus, from the point of view of the production of oxides of nitrogen, it would appear better to use as small a particle as possible, a fact that obviously conflicts with containment costs.

Costs

We shall be hearing much more about the economics of fluidised-bed boilers later in the Conference. Because we have not met any major snags over the previous year which would be costly to solve, I feel confident that fluid-bed combustion can be, at one and the same time, economical and of great amenity value. This does not mean that we should be complacent. On the contrary, because of the many intriguing possibilities, I would like to devote the remainder of my address to a plea for expansion of our programmes.

Three centuries ago Shakespeare wrote:

> "There is a tide in the affairs of men which, taken at the flood, leads on to fortune."
>
> Julius Caesar.

However to be more appropriate to the modern world, with its rapid technological development, the affairs of men ought now to be compared to a breaking wave. Those familiar with surf riding know that the water in a wave is constantly overtaking that ahead of it; similarly, in research and development, new ideas often overtake the old. Thus the timing of the effort to ride the wave is much more critical than at any previous time in history. Shakespeare also knew this:

> "And we must take the current when it serves, or lose our ventures."

I submit that now is the time for us to consider the next round of development of fluidised-bed combustion, so that we need not embark on a protracted course of sequential improvements.

DEVELOPMENT PROSPECTS

The first improvement on current designs would be to raise the top temperature and pressure of the steam cycle. A later paper outlines the economics of the extra tube costs versus the lowering of the heat rate, so I will not go into that here. However, this is not the only way of improving the overall efficiency: we should not ignore combined gas-turbine/steam-turbine cycles, the Field cycle, or even liquid metal cycles.

To the British, efficiency is extremely important because coal-fired power stations will not be able to compete with nuclear energy unless we can lower both fuel costs and capital costs. Figures 2 and 3 show the situation a year ago.

Coal needed something like a 20-percent reduction in overall costs to compete with the new generations of nuclear plants (if they reach their targets). Since 1969, coal costs have risen by 10 percent: further rises seem inevitable. On the other hand, since nuclear prices have gone up (the cost of nuclear power is now about 0.6d/kW), the situation is no worse for coal. If we can relax the specification for coal by adopting fluidised-bed combustion and increase efficiency, the prospects for coal are much better.

Secondary Heat Exchange Beds

The first Central Electricity Generating Board (C. E. G. B.) Research and Development design studies used a second fluidised bed above the combustion bed to act as an inert heat exchanger, using economiser tubes to cool the combustion gases to the air heater inlet temperature of 750°F. When subsequent studies showed that the pressure drop penalty was too great, the idea was abandoned.

Recent data on extended (finned) surface tubing has led to a revival of interest in systems without convective steam or water tubing. The consequently possible plant-layout simplification and heat transfer surface reduction make this an interesting alternative to existing designs. Note that, whereas in convective systems the increased thermal performance of finned tubing is offset by the increased pressure drop, the even better performance of finned tubes in fluidised beds is accompanied by a reduction in pressure drop.

Circulation of Solids

One way of overcoming the startup and control problem is to remove the cooling tubes from the bed and place them in a separate steam raising unit. The heat is transported from the combustor to the tubes by circulation of bed material via fluidised pipelines or ducts. Since the tubes do not cool the bed directly, the bed is brought up to temperature with a comparatively small auxiliary burner with no solids circulating. Partial loads would be controlled by varying the rate of circulation of solids.

This system has several distinct advantages over "normal" fluidised beds:

1. The steam raising unit is very compact and can be manufactured at the factory. For very large stations, the superheater, the reheater, and the evaporators would be separate; for 120-MW units they could be transported (as a unit) by truck to the site.
2. Locating the steam raising units near the steam turbine eliminates the steam pipes and makes it economical to incorporate multiple reheat.
3. Coal feeding can be improved by using rotary distributors or by rotating the solids in the bed, using either air injection or rotary paddles. Another possibility that might be investigated is the combustion of run-of-mine coal at relatively low velocities. Although the larger coal would drop to the bottom of the bed, the continuous movement across the base would prevent the particles settling; there is a good chance that the system would work. We already have experience with bricks moving around circulating beds under the action of the solids, so that there should be little doubt that 1-in. lumps would keep moving. The saving in coal preparation costs would be quite important; so would be the improvement in outage and maintenance costs of the preparation plant.

4. Because we can more or less dictate the temperature of the fluidised solids which any particular tube material contacts, the design of the tubing can be further optimised; corrosion, which might occur using low-temperature tubes in a high-temperature bed of very bad coal ash, could be eliminated by lowering the temperature of the solids in that area of the tube bundle. (This might be particularly relevant when using heavy oil.)
5. Reducing the startup and control problems and increasing the rate at which loads can be accepted or shed will save live steam, normally dumped during these periods.
6. There is a possibility that introducing special heat carriers into the system will improve still further the compactness of the boiler. This improvement of compactness may not amount to very much at low steam temperatures, but could lead to lower maintenance costs and worthwhile cost reductions in high temperature units.
7. Once heat transport is incorporated, it is then only a short step to heat storage.

Heat Storage

Interposing a large hot-solids hopper between the fluidised combustor and the heat exchanger unit makes it feasible to run the combustor for longer periods than dictated by load requirements. Thus all equipment associated with combustion (namely, coal conveyors, coal mixers, bunkers, crushers, feeders, air fans, the combustor itself, the air heater, gas cleaners and precipitators, chimneys, ash handling gear, and the foundations) and civil engineering associated with this equipment can be operated 24 hours a day. This equipment therefore can be rated at only a fraction of the station output, depending on the load factor for which the plant is intended. Although future base loads are expected to be met by nuclear power plants, there will still be a requirement for low-load fossil-fuel stations: this suggestion is worth at least a paper investigation.

If we take an overall power station cost of about £55/kW, the combustion-related costs would be in the region of £22/kW and the fixed cost dependent on electrical capacity would be about £33/kW. Thus the cost of the storage plant, excluding the storage vessel itself, would be:

$$£33 + \left(£22 \times \frac{L}{24}\right), \text{ where L is the load factor.}$$

For an 8-hour daily duty, about £15/kW can be saved, offset by the cost of the storage vessel. Originally, a double-vessel system was envisaged; but this was soon discarded in favour of a compartmented vessel with one empty space: the solids to be transferred between the compartments in rotation. This system was at a cost disadvantage because the compartment walls had to be designed to withstand the hydrostatic pressure of the solids. The latest idea is to sink a single vessel in the ground, separating the hot solids from the cold with a moveable vertical partition. The zones immediately on each side of the partition would be fluidised, with solids pumped from one to the other depending on whether the plant was storing or recuperating heat. It may even be feasible to use natural sandy terrain with the minimum of containment if the water table is well below the level of the store. However, even without such ideal circumstances, it should be possible to construct the storage unit for a cost in the region of £40/cubic yard. This compares favourably with a cost of roughly £12/cubic yard for an insulated and heated heavy oil storage vessel.

For a 4-hour daily duty, a 500-MW station would need 1.3×10^{10} Btu of heat storage, requiring a 200-ft diameter by 22-ft deep vessel, or equivalent. The cost would be £/2kW. Figure 4, showing how the cost of the storage power station would vary for different load duties, indicates that for low daily usage, the storage system is most attractive.

Apart from cost, such a plant would have several advantages over straight sets:

1. If sulphur recovery is incorporated, the extra equipment is also used on a continuous basis with a further saving in capital costs.
2. The stack effluent, exhausted on a 24-hour basis, for a given electrical output would be less objectionable.
3. The station can be brought on line very quickly. It is almost as good as water pump storage, but with the advantage that locating it in the load centres will save electrical transmission charges.
4. The reliability of the station should be improved. Even if some part of the combustion system fails, there is a period of grace during which no loss of capacity results. An improvement of say 5 percent on this account is equivalent to a reduction of a similar 5 percent in capital cost in an integrated network.

Figure 3 shows how improved efficiency and heat storage can help coal in future competion with nuclear energy.

DIVERSIFICATION

Up to now, fluidised-bed combustion has primarily been aimed at coal combustion; but in the last year or so, attention has spread to the burning of heavy oil and (more lately) to gas combustion. Since the work is still mainly directed at large scale power generation, it might pay us to review other applications, both from pollution and economic standpoints.

If we look at the amount of fuel used, we find that less than half goes to the electrical industry. Small-scale use of fuel in factories, domestic heating, and transport add up to a major portion of fuel consumed; in many cases they represent higher sources of pollution per pound of fuel burnt. Could fluidised-bed combustion help in these cases just as we expect it will in large scale production?

For small scale factory furnaces there is every reason to expect that fluidised-bed boilers or furnaces for metal processing can be just as effective in reducing capital costs and pollution. Here, however, I would criticise the attempts which have been made up to now on packaged shell boilers. If we are to exploit best the advantages of fluidisation, we ought not merely to translate previous designs into fluidised versions. We must survey the whole problem and come up with advanced designs which take advantage of all that the new technology can provide. Our aims must be more a efficient plant, a more easily controlled system, better steaming conditions (users have never had the opportunity to buy anything other than low temperature steam boilers), and a plant that can use a wide variety of fuels.

In this context, I would like to stress the future importance of total energy installations. In the past, such installations have rarely been economical because of the high incremental cost of high temperature boilers. Now we can produce a 1100°F by 2000-psi plant for virtually the same cost as for a 212°F system; economics are radically different. The development of new engine designs, mentioned later, could also have a large bearing on this system which everyone agrees is ideal but seldom appears economical.

Domestic Appliances

At first sight, the application of fluidised-bed combustion to gas-fired central heating systems does not appear to be very attractive: current boilers are fairly highly developed, have reasonable efficiencies, and are not bad from a pollution aspect.

The only advantage of fluidised-bed combustion would be a probable reduction in oxides of nitrogen resulting from low temperature combustion. Note that the tendency, if any, is away from the common fishtail burner and toward a more intense (presumably higher temperature) and compact combustion system which may increase pollution from oxides of nitrogen.

Nevertheless, preliminary designs of small gas-fired fluidised-bed combustors which we have undertaken at Aston show that these systems, too, can be very compact and yield better efficiency than current installations. Because these systems can be adapted to burn oil easily and, with the addition of a feed system, could burn fine coal, they might have wide commercial application.

Room Heating

A great deal of money has been spent in the U.K. developing smokeless briquettes. The main disadvantage is high cost: roughly double that of the original fine coal from which they are made.

I often wonder whether it is too late to consider developing small fluidised-bed combustors for room and house heating. There is no problem in developing a 6- or 8-in. combustor; the efficiency, even without flue gas heat recovery, would be above 66 percent. All that would be required would be a suitable coal-hopper/variable-rate feeder and an air blower unit. Pre-mixing the coal with the right amount of limestone could reduce sulphur pollution to a minimum. Such coal could be delivered either by tanker or in pre-packed bags to eliminate much of the labour involved in normal coal fires.

The constantly changing flame pattern is one of the main attractions of an open coal fire; a similar advantage would be possible with a fluidised bed if it were enclosed in a high-temperature-glass tube (these can be made to withstand 1600°F). The constantly changing bubble and particle motion would be a substitute for the coal flames. Economically, one could afford to pay quite a high initial price for such a unit if it could burn cheap coal.

The disposal of the ash would not constitute a severe problem; it might even be possible to wash it away through the house sewage system.

Combustion of Refuse

Attention is now being drawn to better methods of disposing of household refuse. The low combustion temperatures permissible in fluidised-bed combustors, combined with their thermal inertia which would counteract the variability of the combustible constituents, may prove advantageous.

One problem will be how to deal with the large range of size and density of refuse. Light materials such as wood would float on the surface of the bed until burnt and so should present no difficulty. The ideas mentioned earlier for circulating the bed solids may also be of use in keeping larger dense particles in motion until they are completely burnt or rejected.

The major problem would be the effluent caused by burning plastics. We might be able to reduce the chlorine pollution of the atmosphere by using techniques similar to those being used for sulphur recovery.

Vehicles

One of the major pollution problems is transport vehicles. For urban railways, this has been solved by electrification at high capital cost; but for long distance rail transport, trucks, and cars, the problem has not been fully solved. Various devices for cleaning up petrol and diesel exhausts have been proposed; all increase the cost and decrease the output of the power units.

As a consequence, special attempts are being made to produce prime movers with external combustion systems; the two principal ones are steam engines and hot air engines. In both these cases it is extremely likely that fluidised-bed combustion not only could produce a cheaper indirect firing system but also could use cheaper fuels and produce higher temperatures.

One of the major steam engine problems is the contamination of the working fluid with lubricating oil. This oil is difficult to extract and, on passing through the boiler tubes, undergoes cracking and forms carbon deposits. Both the deterioration in internal heating transfer coefficient and the presence of very high temperature combustion gases on the outside of the tubes result in periodic burnout of the tubes. This problem would be completely eliminated in the fluid-bed boiler; with the tubes designed to stand full bed temperature, carbon deposits would only result in loss of output. Currently, some of my undergraduate students at Aston are building a fluid-bed boiler capable of 1300°F by 2000-psi pressure operation. They are also developing a prototype engine, based on a normal car engine with modified valve gear. Fluidised combustion offers similar advantages for other Rankine-cycle working fluids and for hot air engines.

WE HAVE ONLY SCRATCHED THE SURFACE

The above ideas only scratch the surface of where fluidised combustion may lead; other areas are industrial heating furnaces, gas turbines burning ash-laden fuels, combustion of waste gaseous products at refineries and sewage works, and (perhaps ultimately) the "combustion" of nuclear fuels.

Thus we see that there is an enormous development potential, the exploitation of which now seems near at hand.

I believe that the field is so wide that there need be little conflict of interest either among research workers or firms and I trust that in the next few days we can have frank discussions on many of these aspects which will help us all to frame our future programmes. For my part I shall be concentrating on the flow properties of fluidised systems since these have a mixture of academic and practical problems suitable for study in a technological university, but which, nevertheless, could be extremely important in the overall exploitation of our Conference subject.

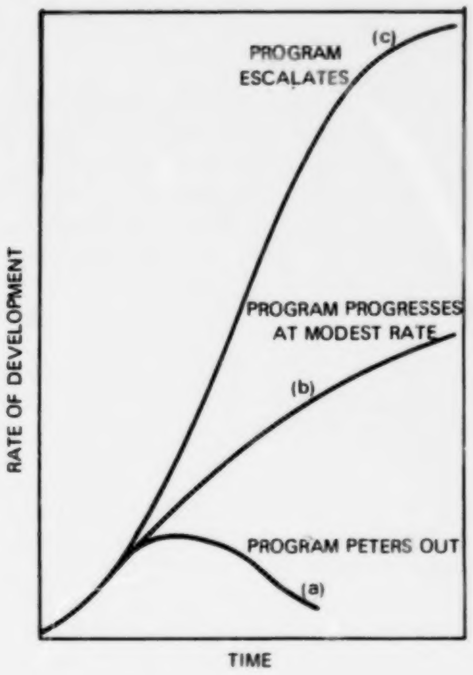

Figure 1. Development of technology.

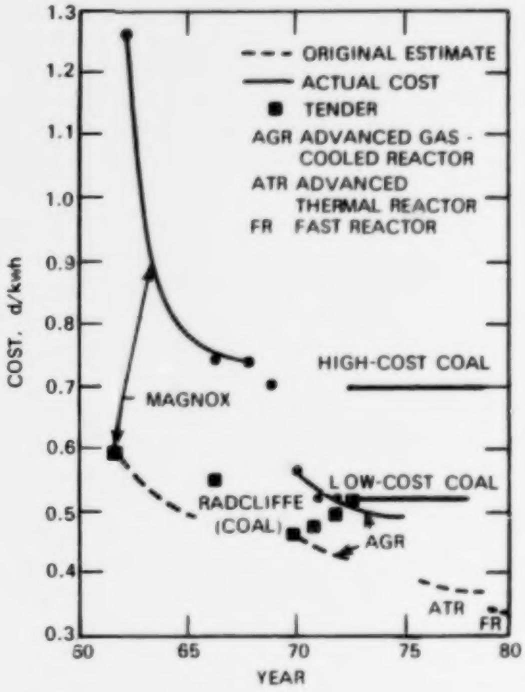

Figure 2. Power cost by type versus year.

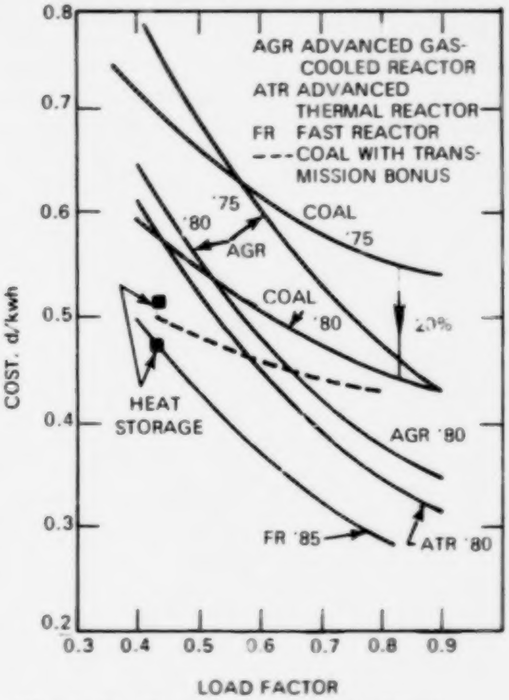

Figure 3. Power cost by type versus load factor.

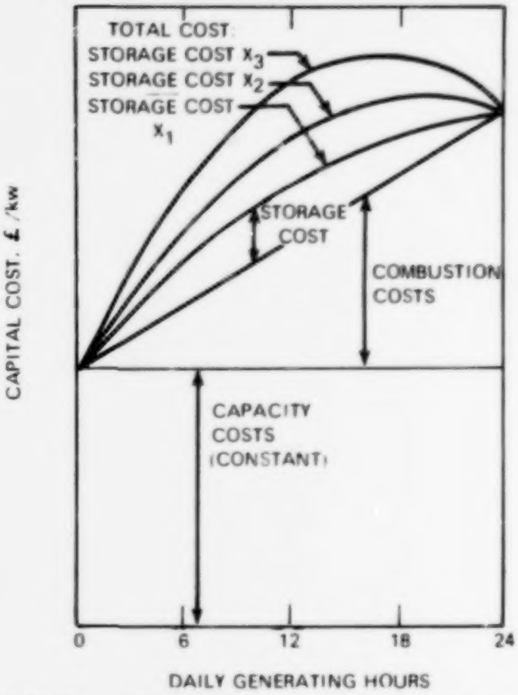

Figure 4. Variation in heat storage cost for different daily load factors.

SESSION I:

Small-Scale Development of Fluidized-Bed Combustion

SESSION CHAIRMAN:

Mr. A. A. Jonke, Argonne National Laboratory

1. PILOT PLANT EXPERIMENTS AT THE COAL RESEARCH ESTABLISHMENT (CRE)

D. F. WILLIAMS

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The purpose of the pilot plant work at the Coal Research Establishment (CRE) has been to obtain data for costing design studies on a 660-MW fluidised-bed boiler, and for the design of a prototype unit. In the preferred concept, crushed coal is fed to a shallow fluidised bed of its own ash, operating within the range of 700 - 900°C. The heat generated is transferred to horizontal tubes immersed in the bed, and the off gases are cooled by conventional means. Elutriated carbon is collected, and is burned by recycling to the original bed or by injection into a separate bed.

At the time of the first Hueston Woods Conference, the initial small-scale work had shown that, at a fluidising velocity of 2 ft/sec, high combustion efficiency could be achieved at moderate temperatures provided that elutriated carbon was recycled to the bed. A high rate of heat transfer between the bed and immersed tubes was demonstrated, and it was found that the rate was not seriously reduced by packing the tubes closely together.

Since that time, a combustor of 3-ft square cross-section has been built to simulate a section of a full-scale bed. For this purpose, it has been provided with insulated walls. The combustor can accommodate full-size tubes in the bed in realistic arrays, which can be changed to test alternative designs. Two of the tubes form part of high pressure water circuit, in which bed-tube heat transfer coefficients can be measured at the tube temperatures that are anticipated for a power station boiler. The remaining tubes are used to extract heat such that the bed temperature can be set at any required level between 700 and 850°C. The bed height is continuously monitored, and is controlled automatically at any level up to 4 ft by withdrawing surplus ash through the base. The type of base and the freeboard height can be varied. Ports in the walls allow pressure and temperature measurements and solid and gas samples to be taken to monitor conditions in and above the bed. Elutriated fines are collected in two cyclones in series, and the primary cyclone fines can be recycled to the bed, either in total or in part, if required.

Runs have been carried out at fluidising velocities between 2 and 8 ft/sec, with coals sized 1/16-in by 0 and 1/8-in. by 0, the width of the combustor having been reduced to 1-1/2 ft to allow the velocity to be increased above 4 ft/sec. The conditions therefore overlap those covered by BCURA in their 27-in. diameter combustor. The coal/air ratio has been varied over wide limits to simulate the conditions that might arise at different distances from a coal feed point in a wide bed. In these runs, carried out without recycling the fines, the rates of combustion in the bed and in the freeboard were determined, together with the amount and size distribution of the unburnt carbon and ash products from the bed, the cyclones, and the gas entering the stack. The efficiency of the cyclones, needed to recover the elutriated fines from a power station boiler without excessive loss of combustible matter, can be calculated from the results. The primary cyclone on the plant was adequate for this purpose, and further experiments were performed to discover how the carbon so collected could be burned. In some of these experiments the primary cyclone fines were continuously recycled to the bed; in others the fines were fed separately to the bed instead of coal to simulate a fines burnup bed.

In each run, the bed size distribution has been measured and has been related to the bed-tube heat transfer coefficient. The effects of tube and bed temperature, bed density, fluidising velocity, and tube packing on the coefficient have also been determined.

A model of what happens to the ash from the time it is injected into the bed as part of the coal feed to the time it is removed as surplus from the bed or is elutriated, including its circulation in the recycle system if this is in use, has been developed. Given the feed rate of the ash, its size distribution in the feed, its resistance to degradation, and the efficiency of the cyclone, the model correlates the combustor data for the quantity withdrawn from the bed, size distribution of the bed, the elutriation rate of each size fraction, the recycle rate, and the change in size through degradation. The model can therefore be used to predict the ability to maintain bed height, and the recycle rate or the rate of feed to a fines burnup bed in a boiler run under a given set of conditions. The model is also of value in predicting the bed size distribution and, hence, the bed-tube heat transfer coefficient.

To find how quickly coal mixes laterally in a bed after injection, measurements of particle mixing rates have been made in a 5-ft diameter cold bed of ash using a radioactive tracer technique. The variables studied include fluidising velocity, particles size, tube packing, and bed height. The results have been incorporated in a mathematical model, based on the combustion and elutriation rates measured in the 3-ft combustor, leading to a prediction of the effect of coal feed point spacing on combustion efficiency. In the model, the carbon concentration gradient between feed points, and the variation in excess air level, are calculated and have been compared with the gradients measured in the combustor.

Another combustor has been built, to investigate the extent to which corrosion, erosion, or deposition may occur on tubes placed in and above the bed. The tubes are made up of different alloys, and are each cooled to a different temperature during the test, which may last for from 100 to 1000 hours. The plant has been designed to run automatically, except for the replenishing of coal feed hoppers, for this length of time.

ACKNOWLEDGMENT

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2. FLUID-BED COMBUSTION OF VARIOUS U.S. COALS

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INTRODUCTION

Fluid-bed combustion has several characteristics which make it potentially attractive as a boiler-firing technique. First, the fluid bed is an efficient contacting medium for gas-solids reactions, thus facilitating reaction of solids with SO_2 to remove it from the combustion products. Also, fluid-bed combustion takes place at lower temperatures than does conventional combustion, and thus should result in lower emissions of oxides of nitrogen. Another potential advantage of the lower combustion temperature is decreased fireside corrosion and deposition. Moreover, lower-grade fuels (high moisture and ash) can be used without certain coal preparation steps that are necessary for conventional pulverized-fuel boilers.

Fluid-bed combustion as a boiler firing technique first received attention in Europe as a method of utilizing anthracite fines[1,2] and lignite[3,4]. Early in the 1960's, experimental programs on fluid-bed combustion were undertaken by England by the National Coal Board (NCB) and British Coal Utilization Research Association (BCURA). NCB has concentrated on development of atmospheric pressure boilers for central station power plants[5]; BCURA has worked on the development of shell boilers[6] and pressurized combustors to be used in a gas turbine cycle[7]. About 5 years ago, research programs were begun in the U.S. by Pope, Evans and Robbins[8,9] and the Bureau of Mines[10,11].

More recently in the U.S., The National Air Pollution Control Administration (NAPCA), through contracts with several organizations, has undertaken a broad research program to investigate fluid-bed combustion because of its potential for reducing emissions of pollutants from fossil-fuel-fired boilers. Under NAPCA contracts, both Argonne National Laboratory[12] and Esso Research and Engineering Company[13] have been investigating the use of limestone to reduce emission of SO_2 from fluid-bed combustors. In 1970, Consolidation Coal Company reported on results of experiments on coal combustion in fluidized beds of dolomite and regeneration of the dolmite[14].

As part of the NAPCA-sponsored program, the Bureau of Mines conducted research to assist evaluation of the technical feasibility of fluid-bed combustion and to obtain engineering data with which to assess its commercial potential. An important consideration in the development of fluid-bed boilers is the amenability of various U.S. coals to this combustion technique. Behavior of the ash may be important, for example. Coal ashes that will remain in the combustion chamber to serve as bed material could be an advantage or a disadvantage, depending on the method of sulfur removal. For instance, if operation with limestone beds is the SO_2-removal scheme, entrainment of ash in the POC (products of combustion) and removal in this manner probably would be preferred. If fine limestone injection is used, retention of the ash as the bed material likely would be preferred. Combustion tests were conducted by the Bureau on a variety of U.S. coals to determine which are most worthy of expanded investigations and to provide a basis for predicting coal types and extent of reserves that would be most suitable for fluid-bed boilers. Results of combustion tests are described in this paper.

EQUIPMENT AND PROCEDURE

Figure 1 shows the 8-foot-high combustor used in the tests. A cone-shaped plate perforated by 1/8-inch-diameter holes supports the fuel bed and admits the fluidizing air; water passing through a heat exchanger of 3/4-inch pipe extracts heat from the combustion bed. Figure 2 is a flow diagram of the system. Coal is fed pneumatically to the base of the fuel bed. Combustion products are passed through two centrifugal separators for removal of most of the entrained solids, and then to a bag filter for further cleaning. Solids from the first separator can be reinjected into the combustion zone. Combustion gas is monitored continuously for O_2, CO_2, and CO. After each test, the residue is removed from the bottom of the combustor by a screw conveyor.

Startup is accomplished in about 2 hours by burning natural gas in the combustor, and then injecting coal or a coal/inert-solids mixture into the combustion chamber. A 2-foot bed is established at about 1200°F, after which the natural gas is shut off and coal or coal/inert-solids are fed at a relatively high rate until the desired bed level is attained. Thereafter, coal is fed at a rate compatible with the designated superficial air velocity.

Originally, coal was introduced by a screw feeder several inches above the air distributor. Even weakly caking coals could not be fed this way, however, because the fuel particles agglomerated before they could mix with the bed material. When fed pneumatically at the bottom, however, even strongly caking fuels (such as Pittsburgh bituminous) could be burned satisfactorily without agglomeration.

COMBUSTION TESTS

Initial objective of the work was to determine if ash from the coal could be kept in the combustion chamber to serve as bed material. In fluid-bed combustion, the fuel volume is much less than the bed volume, hence an inert material (grog) is required. In initial tests with anthracite and bituminous coal, most of the ash was carried out with the combustion products (POC), which kept the ash content of the bed from reaching the desired value (about 99 percent). Operation with a high carbon content bed was possible, but combustion efficiency was poor, as evidenced by excessive amounts of carbon and CO in the POC.

Attempts were made to burn the coal in various inert solids, including sand, sintered fly ash, and sintered bottom ash from a chain-grate stoker, but none of these worked well. Subsequently, mullite, zirconia, and fused alumina were tried and found to be physically stable. Fused alumina was used in most of the tests.

Six coals have been tested, ranging from high-volatile (A) bituminous to low-volatile bituminous. Chemical and size analyses of the coals are given in tables 1 and 2. Results of the combustion tests are given in tables 3 through 6. All tests were made with 1/8-inch x 0 coal.

In the first four tests (runs A-1, B-1, B-2, and C-1), the coals were burned in -16 +48 mesh fused alumina; all components of the system operated satisfactorily.

In test C-2, with high-volatile (B) bituminous coal (Indiana No. 5) in a bed of zirconia, steady-state conditions were maintained for 85 hours without any operating problems.

In tests E-2 and F-2, high-volatile (C) bituminous coal (Central Illinois, No. 6 seam) and medium-volatile bituminous coal (Indiana County, Pa., Lower Kittanning seam) were burned in a bed of -14 +28 mesh alumina. These coals are relatively high in ash, 22 and 24 percent. Ash from both of these coals remained in the bed and had to be periodically removed to maintain a constant bed height. In both tests, recycle rates from No. 1 cyclone apparently were high, creating large temperature differences between the bottom and top of the combustion bed. This is caused by the design of the solids recycle system which is outside the combustor. Recycled solids lose heat in passing through the recycle system, and at high rates of recycle, this has a chilling effect on the lower portion of the combustion bed. This problem was more severe in test F-2, and when a reduction in fluidizing velocity did not sufficiently improve the operation, material from cyclone 1 was not recycled to eliminate the problem. In test E-2, coarser bed material was used to reduce the amounts of material being recycled from the first cyclone.

Tests G-1 and G-2 were made with a low-volatile bituminous coal (Somerset County, Pa.): For G-1, mullite was the bed material; for G-2, Al_2O_3. Test G-1 was made without reinjection of the fines from the primary cyclone, but in G-2 the fines were reinjected. In tables 3 and 4, G-1 shows an excess air of 46.3 percent, which is incorrect owing to a leaking relief valve in the air supply line. This accounts for the high calculated carbon recovery (145 percent) owing to the apparent amount of CO_2 in the POC. Both tests with the low-volatile bituminous coal were terminated early because of difficulty with the combustor. In both cases, large amounts of solids were deposited in the POC lines, causing excessively high backpressures. The reason for this is not yet understood, but it may be important in determining the suitability of coals for fluid-bed combustion. Conventional coal analyses, including ash fusion temperatures, do not appear to adequately describe the suitability of various coals for fluid-bed combustion.

Runs A-2, A-3, A-4-A, and A-4-B were made with Pittsburgh-seam high-volatile (A) bituminous coal from West Virginia, with comparable results (table 4), even with variations in bed height and fluidizing velocity. The large excess air shown for A-2 in tables 3 and 4 was caused by the leak in the air valve and was not actual, whereas the large excess shown in A-4-B was real and was due to malfunction of the oxygen sampling system.

Table 3 shows that carbon utilization is high, usually exceeding 99 percent, except when materials from the first cyclone is not recycled to the bed. In the two cases when fines from the primary cyclone were not recycled to the bed, carbon utilization, at 91 and 86 percent, was unacceptable for commercial operation.

Table 4 gives results of calculations for the theoretical combustion of the coals tested at the same air/coal ratios used in the tests. Actual and theoretical gas analyses, compared in table 3, show generally good agreement except for oxygen content. In the first four tests, the discrepancy was traced to an incorrect span setting of the oxygen analyzer for the range being used. As mentioned previously, the discrepancy in tests G-1 and A-2 resulted from a leaking relief valve in the air supply system.

Table 5 gives mass balances for the inert solids for the combustion tests. For the first four tests (A-1, B-1, B-2, C-1), only an overall weight change is reported and ash behavior is not quantitatively reported. However, it appears that for coal A there was a gradual build-up of ash particles in the bed, and for coals B and C virtually all ash was carried out in the POC. In subsequent tests, more samples were taken to account for all of the inert solids and to permit identification of ash grog components. When washed coals containing less than 14 percent ash were burned and material from the primary cyclone was refined, most of the ash was entrained in the POC. In tests E-2 and F-2, in which high-ash coals were burned, almost half of the ash remained in the bed. This was apparent during the tests because it was necessary to withdraw material from the bed during both tests to maintain a constant bed height.

Mullite, zirconia, and alumina were used successfully as bed materials. Losses of bed material were low, ranging from 0 to 8 pounds, which is a rate of loss of 0.1 percent per hour. The recovery of inert materials ranged from 93 to 100 percent, except for one test. Considering the mass of material to be handled and the many places for solids to accumulate in the system, these values for recovery are acceptable.

Heat transfer in a fluid-bed boiler will be important in establishing the commercial potential of this combustion technique. Hence, overall coefficients were calculated directly from heat transfer data from one U-tube of the heat exchanger in the combustor by the relating $Q = U_o A_o \Delta t$, where

Q = Heat removed, Btu/hr
U_o = Overall heat-transfer coefficient based on outside area of pipe, Btu/hr-ft^2 - °F
A_o = Outside area of pipe, ft^2
Δt = Average temperature difference between water and fluid bed, °F

Overall coefficients, table 5, show very little difference in magnitude when the tube was in the bed or slightly above the bed, the average being about 53 Btu/hr-ft^2 - °F.

Table 6 shows the particle size distribution of the dust from No. 2 cyclone and the filter bags, as determined by a Coulter counter. The cyclone effectively removed the dust down to about 10-15 microns, with the filter bags removing the plus 2-micron particles.

CONCLUSIONS

A fluid-bed combustor was developed and operated successfully with a variety of coals, including highly caking types, under nominal operating conditions of 3 feet per second fluidizing velocity and 1600 °F bed temperature. Mullite, zirconia, and alumina worked satisfactorily as bed materials, and attrition losses were low. Carbon utilization exceeded 99 percent when material from the primary cyclone was refired; without refiring, utilization was approximately 90 percent. When washed coals containing less than 14 percent ash were burned, virtually all of the ash was entrained in the POC. In the case of two coals containing ash in excess of 20 percent, approximately half of the ash remained in the bed, indicating that for these coals, the ash could be the bed material. Overall coefficients of heat transfer from the bed to a water-cooled tube averaged about 53 Btu/hr-ft^2 -°F when the tube was immersed in the bed, approximately the same as when the tube was a few inches above the bed.

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Table 1. ANALYSES OF COALS BURNED IN FLUID-BED COMBUSTION TESTS

Constituent, %	[a,b] A	[a,c] A_1	B	C	E	F	G	[a,d] A_2
Proximate analysis								
Moisture	1.2	1.3	2.3	5.2	1.2	0.9	0.6	1.3
Fixed carbon	53.3	50.0	49.7	46.6	36.5	55.5	67.4	54.1
Volatile matter	36.5	37.5	39.8	38.9	40.2	19.4	18.1	34.2
Ash	9.0	11.2	8.2	9.3	22.1	24.2	13.9	10.4
Ultimate analysis								
Carbon	74.10	73.05	72.27	68.01	51.92	63.83	75.31	73.47
Hydrogen	5.32	4.72	5.54	5.24	4.57	4.11	4.19	5.23
Oxygen	6.13	5.46	6.35	7.00	15.15	2.43	1.97	6.35
Nitrogen	1.27	1.37	1.47	1.32	1.02	0.98	1.23	1.47
Sulfur	2.98	2.90	3.87	3.99	4.06	3.50	2.83	1.80
Heating value (Btu/lb)		12,260	12,930	11,940	9,290	11,300	13,085	–
[j]Major constituents of ash								
SiO_2	44.1	45.9	38.5	42.3	50.4	47.6	44.3	43.7
Al_2O_3	23.0	24.3	19.5	19.5	19.5	27.4	25.9	21.3
Fe_2O_3	18.3	15.8	–	28.0	15.5	16.5	21.7	19.3
Alkali metals in ash								
P_2O_3	1.0	–	–	0.9	–	–	–	–
TiO_2	1.8	–	–	1.3	–	–	–	–
CaO	5.1	–	–	2.9	–	–	–	–
MgO	1.9	–	–	1.3	–	–	–	–
NaO	1.1	0.8	–	0.6	1.4	0.2	0.3	0.7
K_2O	1.6	2.0	–	2.1	1.8	1.4	1.7	1.7

[a] High-volatile (A) bituminous, Pittsburgh bed, West Virginia

[b] Analysis A applies to Run A-1.

[c] Analysis A_1 applies to Run A-2.

[d] Analysis A_2 applies to Runs A-3, A-4-A, A-4-B.

[e] High-volatile (A) bituminous, Pittsburgh bed, Ohio

[f] High-volatile (B) bituminous, Indiana No. 5 seam, Indiana

[g] High-volatile (C) bituminous, Central Illinois No. 6 seam, Illinois

[h] Medium-volatile bituminous, Lower Kittanning seam, Pennsylvania

[i] Low-volatile bituminous, Upper Kittanning seam, Pennsylvania

[j] Analysis of material ashed at 800° C.

Table 2. SIZE ANALYSIS OF COALS BURNED IN FLUID-BED COMBUSTION TESTS, WEIGHT-PERCENT

Screen size, mesh	Test				
	E-2	F-2	G-1	A-3 A-4-A A-4-B	G-2
+ 10	26.4	26.3	20.3	23.0	23.0
- 10 + 30	39.4	39.0	40.8	32.5	27.1
- 30 + 60	17.5	17.5	20.5	21.3	24.8
- 60 + 100	4.7	6.0	5.8	7.6	8.1
- 100 + 150	3.1	3.5	3.5	4.1	4.1
- 150 + 200	2.0	1.9	1.8	2.3	2.2
- 200	6.6	5.8	5.5	8.8	8.0

Table 3. RESULTS OF FLUID-BED COMBUSTION TESTS OF 1/8-INCH x O COALS

	Run A-1	Run B-1	Run B-2	Run C-1
Bed size, U.S. sieve	-16 +48	-16 + 48	-16 + 48	-16 +48
Bed material	Al_2O_3	Al_2O_3	Al_2O_3	Al_2O_3
Duration, hr	69	89	77.5	30
Avg bed temp, °F	1450	1650	1635	1635
Bed height, in.	20	20	20	20
Superficial vel, ft/sec	2.8	3.0	3.0	3.2
Coal fed, lb	2296	2706	2223	964
Avg coal rate, lb/hr	33.3	30.4	28.7	32.1
Avg air/coal, scf/lb	145	154	174	160
Theor air/coal, scf/lb	133	132	132	124
Excess air, pct	8.6	17.1	31.6	29.2
POC comp, pct				
CO_2	15.2-16.2	14.5-16.0	15.0-16.0	15.0-15.6
CO	0.0-0.5	0.0-0.3	0.0-0.1	0.0-0.1
O_2	0.0-0.3	0.0-0.5	2.0-3.0	1.1-2.0
SO_2	0.16	0.32	0.33	N.D.
Carbon, lb				
In coal	1702	1956	1606	766
From cyclone	31	11	5.8	1.1
From filter	21	7.5	2.8	1.3
In POC	1586	1919	1818	720
Carbon recovery, pct	96.2	99.1	113.7	94.3
Carbon utiliz, pct	96.9	99.1	99.5	99.7
Sulfur, lb				
In coal	68.4	104.7	86.0	38.5
Cyclone solids	2.0	0.3	0.1	0.3
Filter solids	2.2	0.6	0.7	0.4
Bed residue	0.3	0.1	0.1	0.2
SO_2 in POC, ppm				
By difference	2420	2930	2735	2900
From gas analysis	1600	3200	3300	N.D.
Oxygen, lb				
In coal	141	172	141	67
In air	5903	7423	6858	2740
Total input	6044	7595	6999	2807
In POC (CO_2, O_2, CO, SO_2)	4278	5283	5723	2103
As water vapor	977	1199	986	
Total output	5255	6482	6709	2507
Oxygen accounted for, pct	86.9	85.3	95.9	89.3

Table 3 (continued). RESULTS OF FLUID-BED COMBUSTION TESTS OF 1/8-INCH x O COALS

	Run C-2	Run E-2	Run F-2[a]	Run G-1[a]
Bed size, U.S. sieve	-14 +28	-8 +28	-14 +50	-16 +50
Bed material	ZrO_2	Al_2O_3	Mu & Al_2O_3	Mullite
Duration, hr	85	56	57	24.7
Avg bed temp, °F	1600	1488	1543	1555
Bed height, in.	24	14	14	16
Superficial vel, ft/sec	3.0		2.4	3.4
Coal fed, lb	2121	2679	1700	711
Avg coal rate, lb/hr	25.0	47.8	29.8	28.8
Avg air/coal, scf/lb	195	103	135	194
Theor air/coal, scf/lb	124	92	115	132
Excess air, pct	57.0	12.1	17.7	46.3
POC comp, pct				
CO_2	12.45	15.6	15.0	17.1
CO	0.0	0.0	0.0	0.0
O_2	5.0	2.0	2.2	1.0
SO_2	0.25	0.8	0.5	0.22
Carbon, lb				
In coal	1443	1391	1085	535
From cyclone	2	17	87	71
From filter	1	6	6	4
In POC	1514	1266	1018	701
Carbon recovery, pct	105.1	92.7	102.4	145.0
Carbon utiliz, pct	99.8	98.3	91.4	86.0
Sulfur, lb				
In coal	84.6	108.7	59.5	20.1
Cyclone solids	0.7	4.1	4.4	0.8
Filter solids	1.6	1.8	0.2	0.2
Bed residue	6.6	2.1	2.4	–
SO_2 in POC, ppm				
By difference	2940	5820	3820	2220
From gas analysis	2500	8000	5000	2200
Oxygen, lb				
In coal	149	406	41	14
In air	7338	4925	4088	2448
Total input	7487	5331	4129	2462
In POC (CO_2, O_2, CO, SO_2)	5739	3981	3204	2004
As water vapor	889	979	559	233
Total output	6628	4960	3763	2242
Oxygen accounted for, pct	88.5	93.0	91.1	91.0

[a] Without recycle from first cyclone.

Table 3 (continued). RESULTS OF FLUID-BED COMBUSTION TESTS OF 1/8-INCH x O COALS

	Run A-2	Run A-3	Run A-4-A	Run A-4-B	Run G-2
Bed size, U.S. sieve	-14 +28	-14 +28	-14 +28	-14 +28	-14 +28
Bed material	Al_2O_3	Al_2O_3	Al_2O_3	Al_2O_3	Al_2O_3
Duration, hr	73	42	71	24	20.5
Avg bed temp, °F	1569	1606	1571	1585	1576
Bed height, in.	18	24	16	16	18
Superficial vel, ft/sec	3.1	2.6	2.9	3.1	3.0
Coal fed, lb	1966	1210	2289	652	604
Avg coal rate, lb/hr	26.9	28.8	32.2	27.6	29.4
Avg air/coal, scf/lb	186	145	145	181	165
Theor air/coal, scf/lb	129	131	131	131	132
Excess air, pct	43.8	10.8	10.2	37.6	24.8
POC comp, pct					
CO_2	15.4	16.7	16.2	16.4	16.4
CO	0.0	0.0	0.0	0.0	0.0
O_2	1.3	2.2	2.0	2.0	2.4
SO_2	0.16	0.16[a]	0.13	0.08	0.16
Carbon, lb					
In coal	1436	889	1682	487	455
From cyclone	6.7	4.7	9.6	2.3	3.2
From filter	3.5	2.1	4.8	1.5	2.6
In POC	1645	879	1596	583	491
Carbon recovery, pct	115.3	99.6	95.8	120.7	109.2
Carbon utiliz, pct	99.3	99.2	99.1	99.2	98.7
Sulfur, lb					
In coal	57.0	21.8	41.2	11.9	17.1
Cyclone solids	1.3	0.7	1.5	0.5	0.1
Filter solids	1.2	0.6	1.4	0.5	0.1
Bed residue	0.5	–	–	–	–
SO_2 in POC, ppm					
By difference	2361	1860	1850	1460	2700
From gas analysis	1600	1600	1270	800	–
Oxygen, lb					
In coal	107	77	145	42	12
In air	6506	3126	5888	2126	1773
Total input	6613	3203	6033	2168	1785
In POC (CO_2, O_2, CO, SO_2)	4805	2675	4819	1754	1513
As water vapor	742	506	958	277	202
Total output	5547	3181	5777	2031	1715
Oxygen accounted for, pct	83.9	99.3	95.8	93	96

[a] From Run A-2 for calculations.

Table 4. COMPARISON OF ACTUAL POC ANALYSES WITH ANALYSES CALCULATED FOR COMPLETE COMBUSTION AT SAME AIR/COAL RATIOS

POC	A-1		B-1		B-2		C-1	
comp, %	Theor	Actual	Theor	Actual	Theor	Actual	Theor	Actual
CO_2	16.6	15.2-16.2	15.2	14.5-16.0	13.5	15.0-16.0	13.7	15.0-15.6
CO	0.0	0.0-0.5	0.0	0.0-0.3	0.0	0.0-0.1	0.0	0.0-0.1
O_2	1.7	0.0-0.3	3.1	0.0-0.5	5.2	2.0-3.0	4.9	1.1-2.0
N_2	81.4	–	81.4	–	81.1	–	81.10	–
SO_2, ppm[a]	2400	2420	3000	2930	2700	2735	3025	2900

POC	C-2		E-2		F-2		G-1		A-2	
comp, %	Theor	Actual	Theor	Actual	Theor	Actual	Theor	Actual	Theor	Actual
CO_2	11.2	12.4	16.1	15.6	15.2	15.0	12.4	17.1	12.6	15.4
CO	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
O_2	7.8	5.0	2.3	2.0	3.2	2.2	6.7	1.0	6.5	1.3
N_2	80.8	82.3	81.1	81.6	81.2	82.3	80.6	81.7	80.7	83.1
SO_2, ppm[a]	2470	2940	4730	5820	3130	3820	1750	2220	1870	2360

POC	A-3		A-4-A		A-4-B		G-2	
comp, %	Theor	Actual	Theor	Actual	Theor	Actual	Theor	Actual
CO_2	16.3	16.7	16.4	16.2	13.6	16.4	14.6	16.4
CO	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
O_2	2.1	2.2	2.0	2.0	5.8	2.0	4.3	2.4
N_2	81.5	81.1	81.5	81.6	81.0	81.5	80.9	81.2
SO_2, ppm[a]	1500	1860	1510	1850	1200	1460	2060	2700

[a] Actual SO_2 calculated from sulfur balance: sulfur in gas = sulfur in coal minus sulfur in recovered solids.

Table 5. MASS BALANCE FOR INERT SOLIDS, LB

Test No.			A-1	B-1	B-2	C-1
Ash in coal, wt-pct			9.0	8.2	8.2	9.3
Bed material			Al_2O_3	Al_2O_3	Al_2O_3	Al_2O_3
Bed size			-16 +48	-16 +48	-16 +48	-16 +48
Heat-up Period:						
	Inputs:	Grog	200	200	200	200
		Anthracite ash	–	–	–	–
		Test-coal ash	–	–	–	–
		Total	–	–	–	–
	Output:	From POC	–	–	–	–
	Bed materials end of heat up		–	–	–	–
Test Period:						
	Inputs:	Bed material at start	200	200	200	200
		Coal ash	207	222	183	102
		Total	[a]407	[a]422	[a]383	[a]302
	Output:	Cyclone	98	64	149	41
		Bag filter	101	79		37
		Bed withdrawals	0	0	0	0
		Bed residue	275	195	197	218
		Total	474	338	346	296
	Net weight change in bed:[b]		+75	- 5	- 3	+18
	Grog lost from bed:		–	–	–	–
	Ash retained in bed:		–	–	–	–
Ave. Overall Heat Trans Coefficient, Btu/hr/ft^2/°F			[c]67	[c]58	[c]56	[c]54

[a] Does not include ash added during heat-up, but this amount is probably 10% or less of ash input.

[b] Material withdrawn from bed during test is considered as bed residue for this computation.

[c] Heat exchanger tube in bed.

Test No.		C-2	E-2	F-2	G-1	A-2	A-3	A-4-A	A-4-B	G-2
Ash in coal, wt-pct		9.3	22.1	24.2	13.9	11.2	10.4	10.4	10.4	13.9
Bed material		ZrO_2	Al_2O_3	Mu+Al_2O_3	Mullite	Al_2O_3	Al_2O_3	Al_2O_3	Al_2O_3	Al_2O_3
Bed Size		-14 +28	-8 +24	-8 +24	-16 +50	-14 +28	-14 +28	-14 +28	-14 +28	-14 +28
Heat-Up Period:										
Inputs:	Grog	260	128	125	150	175	247	150	–	177
	Anthracite ash	18	11	13	16	15	16	16	–	19
	Test-coal ash	16	19	27	102	14	16	18	–	31
	Total	294	158	165	268	204	279	184	–	227
Output:	From POC	33	33	28	126	30	28	37	–	43
Bed materials end of heat up		261	125	137	142	174	251	147	136	184
Test Period:										
Inputs:	Bed material at start	261	125	137	142	174	251	147	136	184
	Coal ash	196	591	411	99	221	136	239	69	84
	Total	457	716	548	241	395	387	386	205	268
Output:	Cyclone	78	238	221	71	75	50	94	30	22
	Bag filter	86	91	29	14	80	49	91	34	24
	Bed withdrawals	0	184	159	–	–	54	65	22	–
	Bed residue	275	158	166	156	211	211	136[b]	162	220
	Total	439	671	575	241	366	364	–	248	266
Net weight change in bed:[a]		+14	+217	+188	+14	+35	+14	–	+48	+36
Grog lost from bed:		6	8	7	3	0	0	1	0	1
Ash retained in bed:		20	225	195	17	35	14	–	48	37
Bed residue size, mesh:										
	+14								21	10
	-14 +28								54	80
	-28								25	10
Ave. Overall Heat Trans Coefficient, Btu/hr/ft^2/°F		[c]38	[d]63	[d]54	[d]52	[e]52	[c]53	[d]51	[d]50	[e]47

[a] Material withdrawn from bed during test is considered as bed residue for this computation.

[b] Calculated bed residue; test A-4-B was a continuation of test A-4-A.

[c] Heat exchanger tube immersed in bed.

[d] Heat exchanger tube 2 to 4 inches above bed surface.

[e] Heat exchanger tube at bed surface.

Table 6. SIZE ANALYSIS (COULTER) OF SOLIDS FROM FLUID-BED COMBUSTION TESTS, PERCENT BY WEIGHT

Particle size, μ	Test No.									
	B-2	C-2	E-3	F-2	G-1	A-2	A-3	A-4-A	A-4-B	G-2
					Cyclone No. 2					
+70			13	20	34					
-70+50			5	15	16					
-50+30			10	30	27					
-30+20	4	1	10	16	11	3	3	4	4	4
-20+15	10	6	13	11	6	8	5	7	8	6
-15+10	36	33	49	8	6	34	28	34	33	29
-10+7	23	24				22	26	27	24	22
-7+5	17	19				18	21	16	19	22
-5+3	9	15				13	15	11	12	13
-3+2	1	2				2	2	1		4
					Bag Filter					
+20			1	3	6		1			
-20+15	1	1	2	6	9	1	0	1	1	1
-15+10	9	6	11	17	24	2	4	5	7	4
-10+7	12	10	13	16	10	5	6	10	9	8
-7+5	15	15	18	17	46	12	11	17	16	19
-5+3	40	45	40	29	4	52	47	54	58	58
-3+2	23	22	15	12		28	31	13	9	9

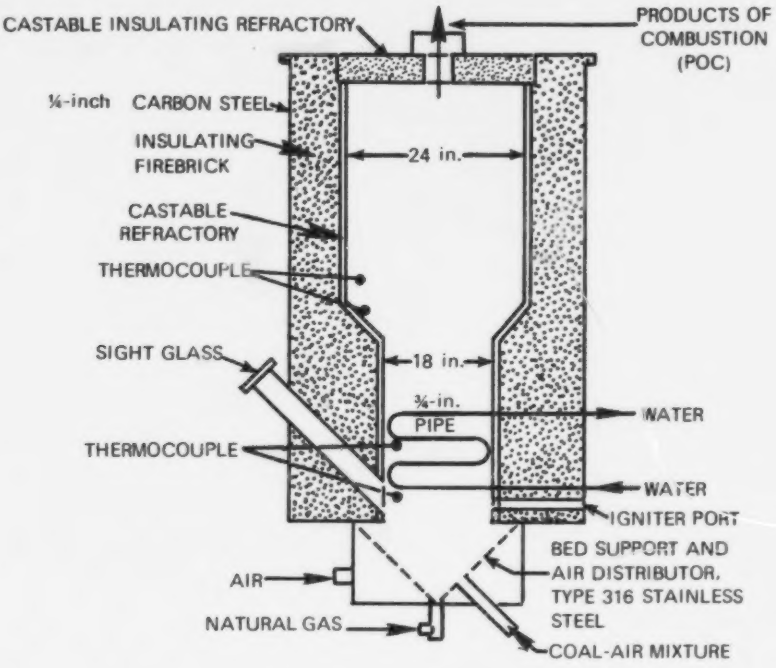
CASTABLE INSULATING REFRACTORY
PRODUCTS OF COMBUSTION (POC)
¼-inch CARBON STEEL
INSULATING FIREBRICK
24 in.
CASTABLE REFRACTORY
THERMOCOUPLE
SIGHT GLASS
18 in.
¾-in. PIPE
WATER
THERMOCOUPLE
WATER
IGNITER PORT
BED SUPPORT AND AIR DISTRIBUTOR, TYPE 316 STAINLESS STEEL
AIR
NATURAL GAS
COAL-AIR MIXTURE

Figure 1. Fluidized-bed combustor.

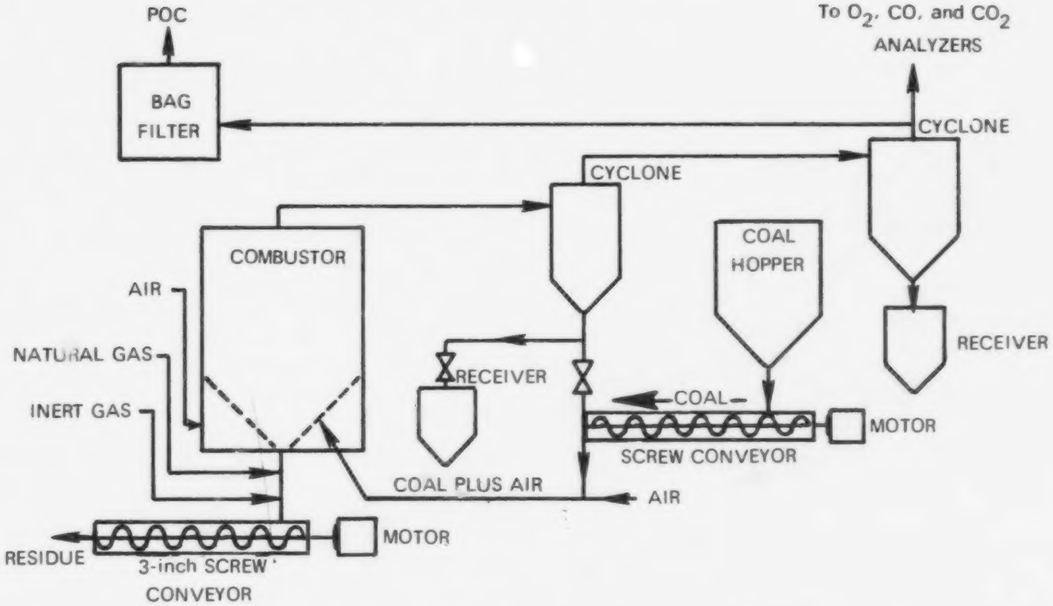
POC
BAG FILTER
To O_2, CO, and CO_2 ANALYZERS
CYCLONE
CYCLONE
COMBUSTOR
COAL HOPPER
RECEIVER
AIR
NATURAL GAS
INERT GAS
RECEIVER
COAL
MOTOR
SCREW CONVEYOR
COAL PLUS AIR
AIR
RESIDUE
3-inch SCREW CONVEYOR
MOTOR

Figure 2. Flowsheet for fluidized-bed combustion system.

3. COMBUSTION OF CARBON-BEARING FLY ASH IN A CARBON-BURNUP CELL

SHELTON EHRLICH

Pope, Evans and Robbins

ABSTRACT

An empirical model which predicts the combustion efficiency for carbon in fly ash in a fluidized-bed combustor is presented. Independent variables considered are fuel feed rate and analysis, air rate, bed temperature, and static bed depth.

The model predicts increasing efficiency with increasing temperature, air rate, and bed depth, but decreasing efficiency with increasing fuel rate and inert matter in the feed.

The model may be used to aid in the design of a carbon-burnup cell.

INTRODUCTION

The National Air Pollution Control Administration (NAPCA), Division of Process Control Engineering, has sponsored work on various applications of fluidized-bed combustion to reduce noxious emissions from power generating plants. One such application is the fluidized-bed boiler in which coal (or possibly residual fuel oil) is burned in a fluidized bed of noncombustible particles. These particles may be selected from a wide range of suitable substances, some of which would be reactive with sulfur.

When coal is the fuel to be fired in a fluidized-bed boiler, an unacceptably high fraction of the energy of the input fuel appears as a high-carbon fly ash. Since a conventional boiler (e.g., pulverized-coal or cyclone-furnace) can achieve combustion efficiency approaching 100 percent, fluidized-bed boilers would not be commercially feasible unless the fuel lost as carbon in fly ash were markedly reduced.

A method proposed by John Bishop[1, 2] involves firing the collected fly ash in an auxiliary combustor. The name "carbon-burnup cell" was coined for this auxiliary combustor.

NAPCA requested that Pope, Evans and Robbins conduct a series of statistically designed tests to model the performance of the carbon-burnup cell. This paper describes these tests and their results.

BACKGROUND

A Description of the Problems

The problem of carbon loss from a coal-fired fluidized-bed combustion device was recognized by Winkler[3] and the fly-coke from Winkler gas generators was used to fire nearby boilers[4]. Even where the combustor was oxidizing, as in Godel's Ignifluid[5], the complete combustion of carbon values could not be achieved in a single pass through the fluidized-bed combustor.

Conventional boilers, using either pulverized fuel or a cyclone furnace, are capable of complete utilization of the fuel in a single pass. The reasons for this performance are clear—the combustion temperature is very high, approaching the adiabatic flame temperature, and the furnace is very large. The reasons for the high conversion efficiency of the conventional boiler lead, however, to certain fundamental deficiencies—large size, high cost, and the inability to practice on-site capture of sulfur—which the fluidized-bed boiler is intended to overcome.

Development of a commercially feasible fluidized-bed boiler, therefore, requires the development of techniques and hardware which would permit a compact and economical boiler to be constructed which would still achieve a high combustion efficiency.

The requirements for combustion efficiency have been understood for some time and are described in a rule-of-thumb by combustion engineers as the three T's—time, temperature, and turbulence. A fluidized-bed boiler in its simplest form cannot meet all of these requirements.

A number of means have been proposed for achieving the desired degree of combustion efficiency: increasing the time factor by recycling; or increasing the temperature factor by operation in an agglomerating mode. Turbulence has already been maximized by use of the fluidized bed and attempts to increase this factor would not be fruitful. However, the increase in excess air is similar in effect to turbulence, causing more oxygen to be made available at the particle surface.

While it was known that combustion efficiency would be favored by high temperature, deep beds (or very low superficial velocities), and high excess air, nothing was known of the relative importance of these variables or how other factors might influence combustion.

A series of statistically designed experiments might answer some of the questions about the performance and design of a carbon-burnup cell. Certainly the system was too complex to treat analytically. A statistical approach could minimize the number of experiments and maximize the information. To this end, and with the aid of a consultant skilled in the design of experiments and their interpretation, we set out to design a series of experiments.

We had some difficulty in finding an individual who possessed the necessary training and experience and who was also capable of relating to engineers and the realities of an engineering apparatus. We did find Professor Arthur Hoerl of the University of Delaware, who did meet our requirements. Much of our understanding of the carbon-burnup process is due to Professor Hoerl's skill.

The Variables

An important first task in the program was to list all of the potential variables and then to categorize them as independent (or control variables), dependent (or response variables), or design variables. It has been clear for some time that certain important variables which have previously been categorized as control variables are, in fact, responses, and so one purpose of this paper is to set this matter straight.

Tables 1 through 6 identify and categorize these variables. Note that the division of the variables into six groups is arbitrary—all control and design variables will influence all responses.

Table 1. COMBUSTION AIR FEED VARIABLES

A. Control variables
  1. Mass flow rate, lb/hr ft^2
  2. Temperature above ambient, °F
  3. Pressure, atmospheres
  4. Composition (e.g., % oxygen and % humidity)

B. Design variables
  1. Distributor design
  2. Injection point of secondary air
  3. Arrangement of secondary injection system

C. Response variables
  None

Table 2. FUEL FEED VARIABLES

A. Control variables

1. Mass flow rate, lb/hr ft^2
2. Temperature above ambient, °F
3. Composition (e.g., % carbon, % inert diluent, and % moisture)
4. Particle size distribution

B. Design variables

1. Injector design
2. Injector location
3. Recycle rate

C. Response variables

None

D. Natural material variables

1. Ignition temperature
2. Porosity
3. Ash particle size
4. Ash sintering temperature
5. Degree of graphitization of carbon
6. Ash/carbon matrix characteristics
7. Attrition characteristics
8. Swelling characteristics
9. Surface area
10. Shape factor
11. Density
12. Emissivity
13. Diffusivity

Table 3. BED VARIABLES

A. Control variables

1. Mass, pounds (weight)/square toot of grid area
2. Static depth, ft
3. Composition (e.g., nominal particle size, density, thermal properties, and hardness)
4. Temperature history (e.g., is the steady state temperature approached from a higher or lower initial temperature)

B. Design variables

1. Combustor shape (e.g., constant or conical cross section)
2. Internals (e.g., arrangement of heat transfer surfaces and baffle screen)
3. Circulation of hot or cold particles
4. Heat sink (e.g., surface area, effectiveness, and temperature)
5. Solids removal method
6. Cross-sectional area, ft^2

C. Response variables

1. Nominal or average bed temperature, °F
2. Bed temperature distribution; e.g., T(x,y,z)
3. Effective thermal conductivity—axial and horizontal, Btu/ft^{2}hr°F/ft
4. Diffusivity—axial and horizontal, ft^2/hr
5. Expanded bed height, ft
6. Bed density distribution; e.g., ρ (z)
7. Bed circulation patterns
8. Equilibrium particle characteristics (e.g., particle size and shape)
9. Residence time distribution of fine particles
10. Fines capture rate through agglomeration
11. Ash particle size increases through agglomeration
12. Viscosity of bed

Table 4. COMBUSTION-GAS STREAM VARIABLES

A. Control variables

None

B. Design variables

1. Arrangement of above-bed heat transfer surfaces

C. Response variables

1. Mass flow rate, lb/hr (or superficial velocity ft/sec)
2. Final composition (e.g., % O_2, N_2, CO_2, CO, H_xC_y, SO_x, NO_x, and H_2O)
3. Composition distribution; e.g., O_2(x,y,z) and SO_2(x,y,z)
4. Temperature distribution
5. Final or average temperature, °F
6. Electrical properties (for electrostatic precipitation)
7. Solids loading, grains/scf

Table 5. SOLIDS STREAM VARIABLES

A. Control variables

None

B. Design variables

1. Freeboard (disengaging height)
2. Above-bed baffles
3. Secondary air injection

C. Response variables

1. Mass flow rate, lb/hr
2. Final composition (e.g., % C, H_2, S, N_2, Cl, and Na+K)
3. Size and density distribution
4. Resistivity (for electrostatic precipitation)
5. Carbon content vs size distribution

Table 6. REACTOR VARIABLES

A. Control variables

1. Position of cooling coils
2. Coolant temperature

B. Design variables

(Covered under fuel, air, bed, and products of combustion)

C. Response variables

1. Steam, generated or superheated
2. Erosion of surfaces
3. Corrosion of surfaces
4. Slagging of surfaces

Variables Actually Studied

While the listing (Tables 1 through 6) is extensive, it cannot include all of the variables; it would have been impossible even to attempt to study all of those listed.

It was necessary, therefore, to select a limited number of variables which would allow some understanding of the process and hopefully answer the basic question—will a carbon-burnup cell work?

Table 7, below, lists the control variables selected, the range suggested at the start of the study, and the range actually investigated. Note that for the purposes of a test program, temperature, excess air, and superficial velocity are considered control variables despite the fact that they are responses.

Table 7. CONTROL VARIABLES SELECTED FOR CARBON-BURNUP CELL INVESTIGATION

Variable	Range Initially Suggested	Range Actually Studied
1. Carbon content of fly ash, %	20 to 80	23 to 65
2. Particle size of fuel	As received and 70% through 200 mesh	As received
3. Static bed depth, in.	6 to 18	10 to 22
4. Nominal bed temperature, °F	1500 to 2100	1700 to 2150
5. Excess air, %	0 to 50	10 to 90
6. Superficial velocity, fps	5 to 20	6 to 15.3

In addition to the control variables indicated in Table 7, it was found desirable to add some special experiments which might point out design considerations. These special tests included the addition of coal and operation with two bed particle-size distributions.

The effect of coal addition and particle size could not be included in the modeling process. Neither was found to be of major importance over the range studied.

Table 8 lists the data for those tests which were useful in the model.

Table 8. EXPERIMENTAL TEST CONDITIONS AND RESULTS

Run No.	Bed Temp, °F	Air Rate, lb/hr	Bed Depth, in.	Carbon Rate, lb/hr	Inert Rate, lb/hr	Part. Size[a]	Cooling Coil	CE,[b] %	CO_2, %	O_2, %	SO_2, ppm	NO, ppm
C-302-1	1800	760	18	40	33	+		90	11.5	9.1	350	560
-2	2140	760	18	53	52	+		83	14.2	6.4	800	670
-3	1780	820	18	44	28	+		86	10.8	9.7	550	570
C-303-1	1750	330	18	21	20	+		65	9.8	10.8	320	470
-2	1980	330	18	24	22	+		66	11.5	8.0	600	480
-3	1750	500	18	39	29	+		65	12.0	8.5	320	410
-4	1860	500	18	37	30	+		74	13.2	7.4	500	570
-5	1970	500	18	39	32	+		78	14.5	6.2	700	600
C-306-1	2100	600	18	41	65	–		86	14.3	6.2	800	720
-2	1750	860	18	56	78	–		70	11.0	9.3	750	560
-3	2120	775	18	53	77	–		89	14.6	5.8	800	770
-4	1800	600	18	43	62	–		73	12.4	8.2	750	520
C-307-1	1800	600	17	41	65	+		76	13.2	7.5	500	410
-2	2100	330	18	26	39	+		90	16.8	3.5	600	500
-3	2140	600	18	44	66	+		91	16.0	4.8	650	550
-4	1750	330	18	29	44	+		63	13.4	6.9	700	370
C-308-1	1900	600	18	38	49	+		81.5	13.4	7.1	600	520
-2	1900	600	18	42	58	+		81.3	14.5	6.2	750	500
-3	1900	600	18	43	59	+		80.6	15.0	5.5	800	520
C-310-1	2040	700	18	46	59	+		84.6	13.2	7.1	700	460
-2	2000	700	18	55	70	+	IN	75.8	13.8	6.4	720	420
C-311-1	2000	700	18	52	106	+		77.0	16.0	4.5	700	570
-2	1930	700	18	63	127	+	IN	66.7	16.6	3.6	800	470
-3	2000	600	18	79	171	+	IN	58.0	18.1	2.1	850	470
-4	1980	600	18	51	107	+		75.8	15.9	4.5	820	500
-5	1950	800	18	75	157	+		64.5	15.8	4.7	(820)	510
-6	1800	800	18	96	206	+	IN	56.2	16.4	3.9	(820)	350
C-312-1	2000	700	18	47	93	+		84.2	13.8	7.0	900	500
-2	1990	700	14	46	92	+		83.8	13.5	7.2	900	480
-3	1980	700	10	51	105	+		76.0	13.2	7.5	900	480
C-313-1	1980	600	18	37	26	+		91.0	13.3	7.5	1160	630
-2	1980	700	18	43	31	+		87.2	12.9	8.0	1160	610
-3	1980	800	18	48	36	+		86.0	12.8	8.1	1160	600
C-314-1	1980	600	10	39	31	+		84.8	13.4	7.4	1050	560
-2	1960	700	10	46	36	+		82.0	13.0	7.6	1050	560
-3	1980	800	10	57	45	+		76.0	13.0	7.8	1050	560
C-315-1	2010	600	18	46	35	+	IN	83.3	15.4	5.5	1250	560
-2	2000	700	18	49	37	+	IN	84.5	15.0	5.8	1250	560
-3	1980	800	18	52	40	+	IN	85.2	14.6	6.0	1250	540

Table 8 (continued). EXPERIMENTAL TEST CONDITIONS AND RESULTS

Run No.	Bed Temp, °F	Air Rate, lb/hr	Bed Depth, in.	Carbon Rate, lb/hr	Inert Rate, lb/hr	Part. Size[a]	Cooling Coil	CE[b], %	CO_2, %	O_2, %	SO_2, ppm	NO, ppm
C-316-1	2000	600	10	50	36	+	IN	74.5	14.8	5.8	1250	520
-2	1990	700	10	58	36	+	IN	76.5	14.3	6.0	1300	510
-3	1980	800	10	63	46	+	IN	72.0	13.8	6.4	1250	520
C-317-1	1980	700	22	52	43	-		82.2	15.5	4.8	1200	570
-2	1980	700	18	51	43	-		81.5	15.3	5.3	1275	570
C-318-1	1900	700	22	61	43	+		72.5	15.8	5.0	1050	550
-2	1950	700	18	56	41	+		79.8	16.0	4.5	1050	550

[a] + = -16 +22 Tyler mesh
- = - 8 +16 Tyler mesh

[b] Combustion efficiency

RESULTS

For the purpose of modeling, the control (or independent) variables were slightly rearranged and included the following:

Variable	In
Bed temperature	°F
Air rate	lb/hr
Static bed depth	in.
Carbon rate	lb/hr
Inert rate	lb/hr

These independent variables were considered in various alternative regression modes.

Empirical regression models of the type

$$y = b_o + b_1x_1 + \ldots + b_kx_k$$

were successively fitted to the data and evaluated through their corresponding F-ratios. the F-ratio is a statistical measure of the variance in the response data which is explained by the model divided by the residence or unexplained variance. If there were no relationship between the independent variables and the response, the F-ratio would tend toward unity. Therefore, the higher the F-ratio, the more reliable the model is in explaining the trends in the data.

The inclusion of bed temperature as an independent variable is unusual: it should be a response to air rate, feed rate, and percent carbon. However, a satisfactory model could not be developed without including bed temperature as a pseudo-independent variable.

The fly-ash feed variable was examined in two ways:

Group 1	Fly ash feed rate,	lb/hr
	Carbon content,	%
Group 2	Carbon feed rate,	lb/hr
	Inert feed rate,	lb/hr

Although these two groupings are identical, it was found that using the cross-product of carbon-feed rate times inert-feed rate improved the reliability of the model.

Various alternative forms were used for representing the combustion efficiency (CE):

1. CE.
2. Log (1- CE/α).
3. Log CE.

The purpose of this step was to develop a response form consistent with the physical case; i.e., combustion efficiency cannot exceed 100 percent. The form

$$\text{Log}\,(1 - CE/\alpha)$$

with α = 95 to 100 percent would be the preferred form. Table 9 is a summary of the

F-ratio for CE, Log (CE), and Log (1-CE/α), with various values of α.

Table 9. F-RATIO FOR VARIOUS FORMS OF REPRESENTATIVE COMBUSTION EFFICIENCY

Form of Combustion Efficiency (CE)	F-Ratio
CE	74.5
Log (1-CE/95)	46.2
Log (1-CE/101)	69.2
Log (1-CE/110)	72.1
Log (1-CE/120)	74.2
Log (1-CE/145)	75.1
Log (CE)	73.8

These results show that the simple form, CE, gives the best result except where α takes on unreal values above 120 percent. The form CE was therefore chosen for the response.

The final form of the model selected was:

Combustion Efficiency = b_0 + b_1 (bed temperature, °F)
+ b_2 (air rate, lb/hr) + b_3 (bed depth, inches)
+ b_4 (carbon rate, lb/hr) + b_5 (inert rate, lb/hr)
+ b_6 (carbon rate, lb/hr) (inert rate, lb/hr).

Table 10 lists the values for the coefficients b_0 through b_6.

Table 10. REGRESSION COEFFICIENTS FOR COMBUSTION EFFICIENCY MODEL

Term	Designation of Coefficient	Value of Coefficient
Constant	b_0	-13.78
Bed temperature, °F	b_1	0.05193^{-1}
Air rate, hr/lb	b_2	0.04620
Bed depth, in.	b_3	0.3831^{-1}
Carbon rate (CR), hr/lb	b_4	-0.8737
Inert rate (IR), hr/lb	b_5	-0.1905
(CR) (IR), hr^2/lb^2	b_6	0.0027

Table 11 shows the calculated and observed values of combustion efficiency.

Table 11. OBSERVED AND CALCULATED COMBUSTION EFFICIENCY

Run No.	Obsrvd	Calc	Result
C-306-1	86	88.9	-2.9
-2	70	71.7	-1.7
-3	89	89.1	-0.1
-4	73	72.1	0.9
C-307-1	76	73.3	2.7
-2	90	90.0	0.0
-3	91	88.8	2.2
-4	63	69.0	-6.0
C-308-1	81.5	82.0	-0.5
-2	81.3	78.3	3.0
-3	80.6	77.5	3.1
C-310-1	84.6	87.3	-2.7
-2	75.8	78.3	-2.5
C-311-1	77.0	78.6	-1.6
-2	66.7	68.0	-1.3
-3	58.0	59.5	-1.5
-4	75.8	73.4	2.4
-5	64.5	67.7	-3.2
-6	56.2	53.8	2.4
C-312-1	84.2	82.3	1.9
-2	83.8	81.0	2.8
-3	76.0	75.1	0.9
C-313-1	91.0	89.0	2.0
-2	87.2	88.4	-1.2
-3	86.0	88.8	-2.8
C-314-1	84.8	83.9	0.9
-2	82.0	81.6	0.4
-3	76.0	78.4	-2.4
C-315-1	83.3	82.7	0.6
-2	84.5	84.4	0.1
-3	85.2	85.5	-0.3
C-316-1	74.5	76.0	-1.5
-2	76.5	73.8	2.7
-3	72.0	73.8	-1.8
C-317-1	82.2	82.2	0.0
-2	81.5	81.4	0.1
C-318-1	72.5	71.2	1.3
-2	79.8	76.2	3.6

CONCLUSIONS

The results of tests and the form of the model indicate that combustion efficiency is favored by increasing temperature, air rate, and bed depth, and by decreasing fuel feed. These results are consistent with the predictions made at the beginning of the test program, with one exception; i.e., the model predicts increasing efficiency with increasing superficial velocity. This was unexpected and in fact the results tend to show no effect for velocity. Combustion efficiencies of 90 percent can be achieved in burning fly ash from a fluidized-bed boiler in a single pass through a carbon-burnup cell. This would then indicate combustion efficiencies of 99 percent for a commercial boiler which included a carbon-burnup cell. It would be expected that a full-size burnup cell (at least 10 ft^2) would give better performance and that additional recycle of flyash would further improve burnup.

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4. PILOT-PLANT EXPERIMENTS AT BCURA

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INTRODUCTION

The objective of the programme of experimental work at BCURA is to provide data for designing steam generators ranging in size from small industrial units (e.g., 8000 lb/hr) to large central power station units (e.g., 660 MW). A large proportion of the effort has been directed toward pressurised fluidised combustion mainly for combined gas/steam power generating plants. The BCURA research programme is complementary to that in progress at the National Coal Board's Central Research Establishment at Cheltenham.

Since the last conference at Hueston Woods:

1. Research on small industrial steam generators has been taken to the prototype stage.
2. A programme of research to obtain specific data for the design of industrial steam generators has been completed.
3. A pressurised pilot-scale combustor has been operated (initially to assess the suitability of the combustion gases for gas turbines and subsequently to obtain process data for design studies).
4. A major programme of pilot- and laboratory-scale investigations on sulphur oxide retention has begun.

These activities have been supported by "cold" model work and rationalised by mathematical models.

EXPERIMENTAL TECHNIQUES AND EQUIPMENT

The main pilot-scale equipment has been described previously; it comprises:

1. A firetube boiler capable of generating 8000 lb/hr of steam at 100 psi, with a 48-inch diameter cross-section fluid bed.
2. A fully cooled pilot-scale combustor with a 27-inch diameter bed cross-section.
3. A 48 by 24 inch bed cross-section pressure combustor capable of operating at pressures up to 6 atmospheres.

The principal features of the combustors are shown in Figures 1, 2, and 3.

A common feature of all is that a major part of the heat transfer surface is contained in the bed.

A fairly wide range of fuel sizes and fluidising velocities have been investigated. High velocities are attractive from the point of view of minimising containment costs but the higher heat transfer rates result in lower tubing costs; the optimum situation remains to be proved.

INFORMATION OBTAINED

Startup

A number of techniques have been used, ranging from preheating the combustion air to burning gas in a bed with higher-than-normal coal content. The latter technique, one form of which was first reported by Pope, Evans and Robbins, is particularly effective when lighting up a heavily cooled bed. Before adopting this, we did not achieve self-sustained combustion in the prototype boiler. The main snag with this startup system, unless it is carefully applied, is smoke emission; however, a wholly satisfactory technique was evolved before the small industrial boiler programme was completed.

Combustion

The principal loss of combustible material is in the form of solid carbonaceous material elutriated from the bed. As might be expected, losses are greater at high than at low fluidising velocities; better combustion efficiencies are currently attained under the latter conditions. Recirculation of carryover material is essential even at low velocities (e.g., 2 ft/sec) to enable high (99 percent or greater) combustion efficiency to be attained.

Experiments have been carried out so far on 1/16-in. x 0, 1/8-in. x 0, and 1/4-in. x 0 coal; the relevant fluidising velocities for these have been 2 ft/sec (pressure combustor), 4-8 ft/sec, and 10-12 ft/sec, respectively.

The carbon-rich particles of the elutriated material are mainly (though not universally) concentrated in the finer size fractions (e.g., $<50\ \mu m$) and more than one pass through the bed are needed to achieve a reasonable degree of burnout. The amount of material to be recirculated can be large enough to affect the heat balance of the bed.

There are a number of ways in which the problem can be alleviated. In the first place, careful design of the coal preparation equipment can keep the amount of fines (e.g., material smaller than $75\ \mu m$) in the coal feed to a low level. Reducing the amount of fines from about 18 percent to about 8 percent on the pressure combustor has, for example, improved combustion efficiency by two units of percentage.

Classification of the carryover material into the appropriate size ranges, rejecting the mainly ash material, and recycling only the carbon rich stream, is one way of reducing the thermal load on the bed while, at the same time, improving combustion efficiency.

An attractive alternative now being explored is using a separate burnout bed for reacting carbon in the elutriated material. Present indications are that low fluidising velocity and/or higher bed temperatures (e.g., >900°C) are needed to achieve a high degree to burnout of the material elutriated from the combustor operated at high fluidising velocities.

More data is needed before a final choice could be made between recycling elutriated carbon to the main bed or to a separate bed; the solution adopted may well depend upon specific features of the application.

Coal Distribution

The attainment of a high combustion efficiency (and a minimum of combustion above the bed) is also strongly dependent upon rapidly achieving uniform distribution of coal over the cross-section of the bed.

Diffusion of a coal stream admitted at the bottom of the bed is more rapid in a vertical than in a horizontal direction–as might be expected–and from this point of view alone a large number of coal nozzles are needed to ensure good lateral dispersion of the coal in a big fluidising bed.

The extent to which lateral dispersion can be assisted by introducing coal streams at high velocities in a lateral direction has been explored in a 5-ft diameter cold model (situated at the National Coal Board's Central Research Establishment). The rate of mixing was deduced from the distribution of radioactivity in samples extracted from the bed after a pulse of irradiated coal had been fed through a nozzle into the bed.

It was found that mixing could be described in terms of diffusion theory and that, over the limited range of variables investigated, the coefficient of diffusion was proportional to the fluidising velocity and to the momentum of the coal particles. Increasing the momentum of the particles by the use of high velocity air streams was found to be difficult, however, due to a "slip" between the coal and air streams.

In general it is also difficult to increase momentum to an effective extent without also increasing the quantity of transport air. Other experimental work has shown, however, that combustion of coal fines and volatiles above the bed is increased if the quantity of transport air exceeds about 2 percent of the total combustion air.

The general conclusion is that other means need to be found for increasing the amount of lateral diffusion achievable from a single coal inlet point. Work to this end is in progress at BCURA and at CRE.

Measurement of Coal Flow

A number of methods are available for trimming the flow of air/coal mixtures so as to achieve reasonably uniform division of flow between streams travelling along a multitude of pipelines.

The problem is to measure the amount flowing; because there will be a large number of pipelines, an inexpensive device is needed. We have tried a number of systems; experimental work continues, but we are far from being able to say, to within even ± 10 percent, how much coal is flowing in a pipeline. Reliable detection of whether or not coal is flowing, however, can be achieved by modern proprietary ultrasonic devices.

Heat Transfer

The initially predicted high values for heat transfer to surfaces immersed in fluid beds have been confirmed. Horizontal tubes arranged on staggered pitches give the highest values–and correlations (one example is shown in Figure 4) between bed particle size and heat transfer coefficients valid to within ± 15 percent have been evolved.

Higher coefficients are achieved to hot than to cool surfaces but there is more scatter in the data for the former, most probably because of the difficulty of accurately measuring the temperature of metal surfaces at high temperatures.

Higher than expected heat transfer rates were achieved in the prototype boiler mainly because the design was based on measurements made to "cold" tubes. Some tubes had to be cut out to eliminate over-cooling which caused unsatisfactory effects on combustion.

ASH BEHAVIOR

The coals tested have generally had ash contents in the range of 15-25 percent. No difficulties have been experienced with clinkering with bed temperatures maintained below about 950°C.

The ability to maintain bed level depends on the nature of the ash (its friability) as well as the quantity available. An empirical test (evolved by the National Coal Board) has been used to assess the degradability of coal ashes; most of the United Kingdom coals tested so far appear to have ashes with reasonably good resistance to degradation. The penalty of adding material (e.g., coal washery shale) to maintain bed level–where a coal ash was too friable–would not, however, be serious.

In the experimental rigs operated at low fluidising velocities, about 20 percent of the input ash is extracted from the bed; however, at high velocities, most is carried out and normally only enough is retained to maintain bed level.

It has been found that by suitably arranging closely spaced horizontal tube surface at the top of the bed, more ash is retained in the bed—and more combustion occurs within the bed.

Deposits and Corrosion of Surfaces in the Bed

Specimens of boiler superheater alloys have been exposed to the conditions in the pressure combustor bed for several hundred hours. The results have generally been similar to those obtained in the specially designed corrosion tests in a special rig at the NCB's Central Research Establishment.

Suitability of Combustion Gases for Gas Turbines

The exhaust gases from the pressurised combustor at 750-800°C are passed over a static cascade of blades to assess the extent of erosion, corrosion, or deposition. The cascade, a nozzle segment from an industrial gas turbine, has now been in operation for several hundred hours. The dust loading of the gases has generally been about 200 ppm, with most of the dust less than 20 μm in size. Because most of the particles greater than 10 μm are in the form of "platelets," it is very difficult to obtain a realistic size distribution of the ash.

The nozzle blades are lightly coated with an ash layer which appears to reach equilibrium after operating about 20 hours. The layer can be limited to a negligible amount by conventional on-line cleaning methods. No signs of erosion have been observed.

While it is realized that only tests with an actual turbine will provide convincing evidence, experience to date is highly promising.

Part-Load Operation

Operation over a wide load range poses a number of problems in a system where bed temperature is the major factor controlling heat generation in the bed and where the range of temperature over which the bed can be operated is relatively narrow.

The main application envisaged for pressurised fluidised combustion is for a combined gas/steam turbine power plant. In this system the combustion and heat transfer characteristics of the fluid bed have to be matched to the pressure, temperature, and mass flow characteristics of the gas turbines.

Two main methods of controlling heat absorption in the bed have been considered and some features have been investigated using "cold" models.

Reduction of load by shutting off sections of the bed appears feasible for atmospheric pressure combustion; but, until a large combustor has been tested, the full extent of the operating problems will not be known.

It is difficult to achieve satisfactory load control for a combined cycle, using this system; one problem is to maintain the air/fuel ratio at a satisfactory level over the whole range.

The alternative method of controlling heat absorption is to alter the amount of tubing immersed in the fluid bed by changing bed level to match the load demand.

Model work has shown rapid change of bed level to be feasible in principle. Figure 5 indicates that this method can be used to maintain a satisfactory air/fuel ratio over a wide load range.

FUTURE WORK

Sufficient data has been obtained from the pilot plants to enable a major scale-up of the atmospheric process to be faced with confidence. This does not, however, mean that all essential background work has been completed; there is still a need for a selective investigation of such factors as:

1. Coal characteristics.
2. "System response" for designing automatic control systems.
3. Methods for completing combustion of material elutriated from the bed.
4. Alternative and possibly more advanced methods for load control that could be incorporated in the scale-up plant at a later date.

Much of the data obtained for operation at atmospheric pressure is relevant to combustion under pressure. Operation of a large bed at atmospheric pressure is an essential stage in scaling up the pressure process.

Operating experience on the pressure process suggests that there is sufficient technical justification to proceed with the design of the next stage, namely, a plant incorporating a small industrial gas turbine to demonstrate the long term feasibility of running turbines on the gases from this combustion system.

Design studies on the pressure process have also shown the need for further investigation at the present scale of operation of such factors as:

1. Ash removal.
2. Coal distribution.
3. Specific features of the proposed load control system.

Another programme of research recently begun at Leatherhead—sulphur retention by lime in both pressurised and non-pressurised fluid-bed systems—is described in a separate paper.

ACKNOWLEDGMENTS

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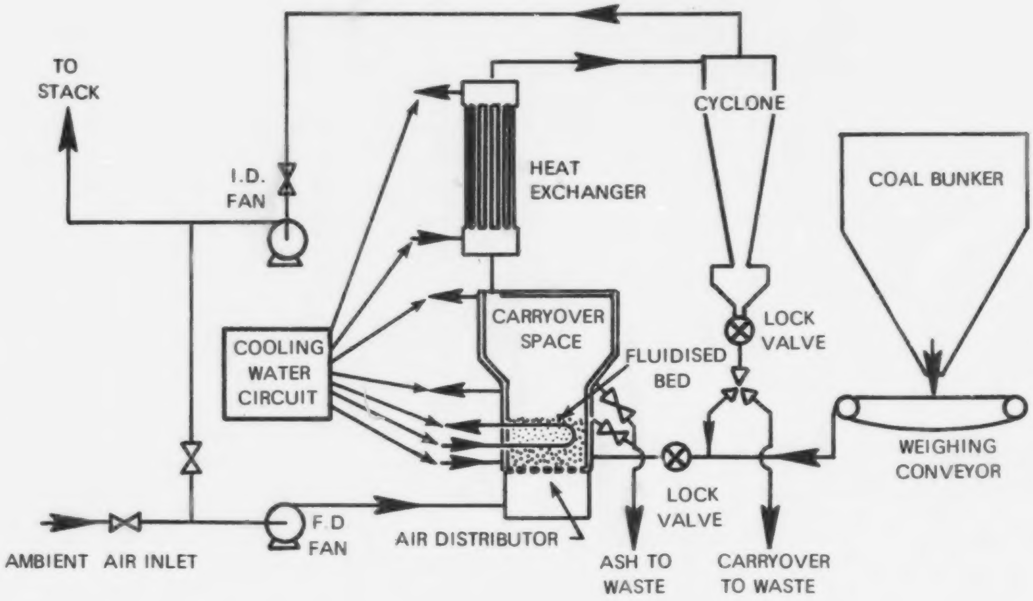

TO STACK
I.D. FAN
HEAT EXCHANGER
CYCLONE
COAL BUNKER
CARRYOVER SPACE
FLUIDISED BED
LOCK VALVE
COOLING WATER CIRCUIT
WEIGHING CONVEYOR
LOCK VALVE
AMBIENT
AIR INLET
F.D FAN
AIR DISTRIBUTOR
ASH TO WASTE
CARRYOVER TO WASTE

Figure 1. 27-inch diameter fluidised-combustion rig.

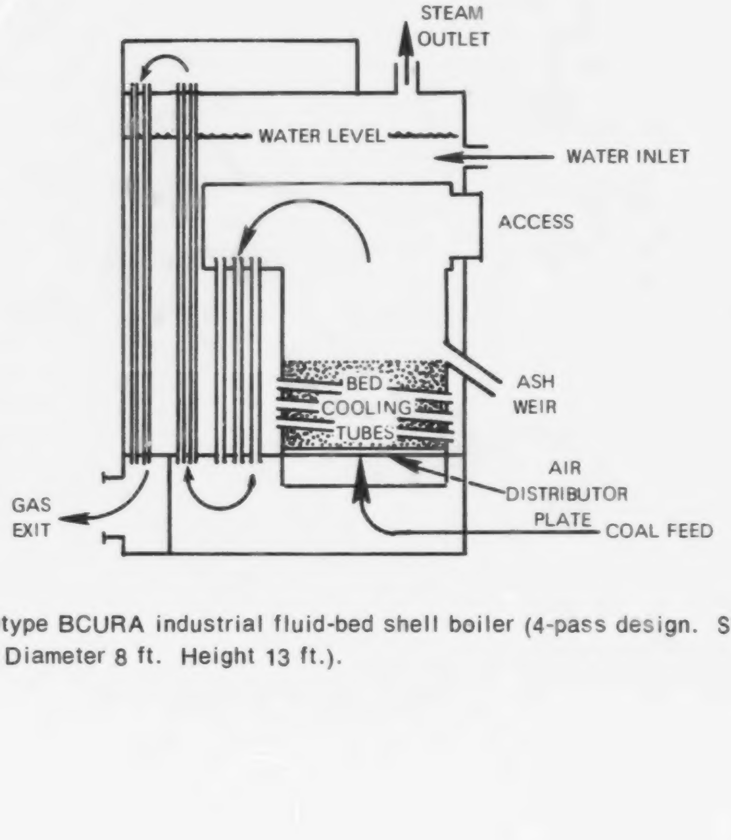
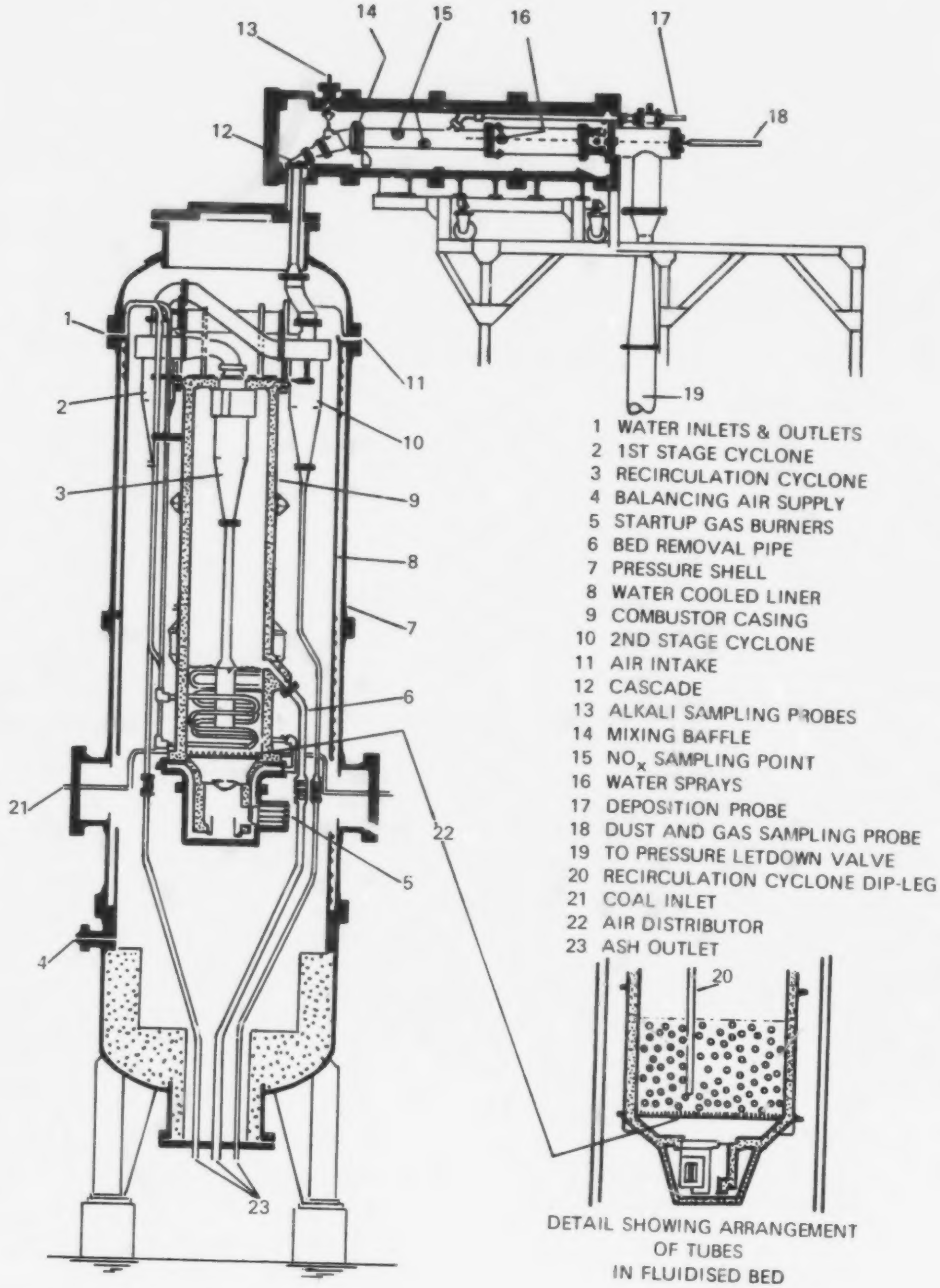

STEAM OUTLET
WATER LEVEL
WATER INLET
ACCESS
ASH WEIR
BED COOLING TUBES
AIR DISTRIBUTOR PLATE
GAS EXIT
COAL FEED

Figure 2. Prototype BCURA industrial fluid-bed shell boiler (4-pass design. Steam output 8000 lb/hr. Diameter 8 ft. Height 13 ft.).



1 WATER INLETS & OUTLETS
2 1ST STAGE CYCLONE
3 RECIRCULATION CYCLONE
4 BALANCING AIR SUPPLY
5 STARTUP GAS BURNERS
6 BED REMOVAL PIPE
7 PRESSURE SHELL
8 WATER COOLED LINER
9 COMBUSTOR CASING
10 2ND STAGE CYCLONE
11 AIR INTAKE
12 CASCADE
13 ALKALI SAMPLING PROBES
14 MIXING BAFFLE
15 NO_x SAMPLING POINT
16 WATER SPRAYS
17 DEPOSITION PROBE
18 DUST AND GAS SAMPLING PROBE
19 TO PRESSURE LETDOWN VALVE
20 RECIRCULATION CYCLONE DIP-LEG
21 COAL INLET
22 AIR DISTRIBUTOR
23 ASH OUTLET

DETAIL SHOWING ARRANGEMENT
OF TUBES
IN FLUIDISED BED

Figure 3. Section through 48'' x 24'' pressurised fluid-bed combustion rig.

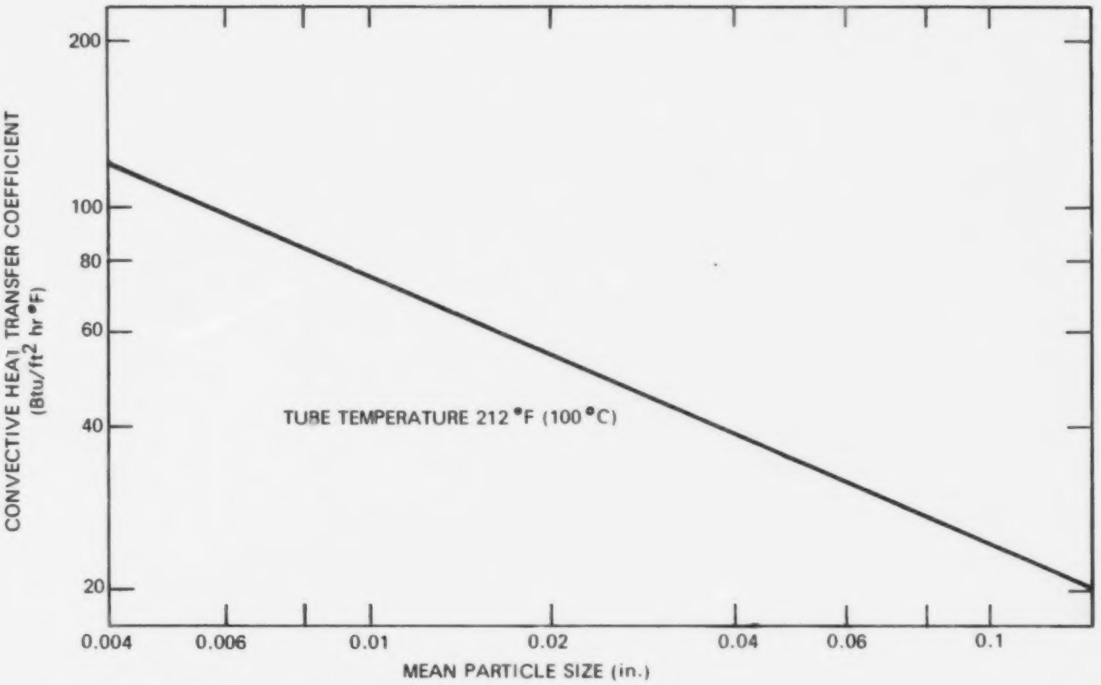


Figure 4. Fluid-bed heat transfer coefficients.

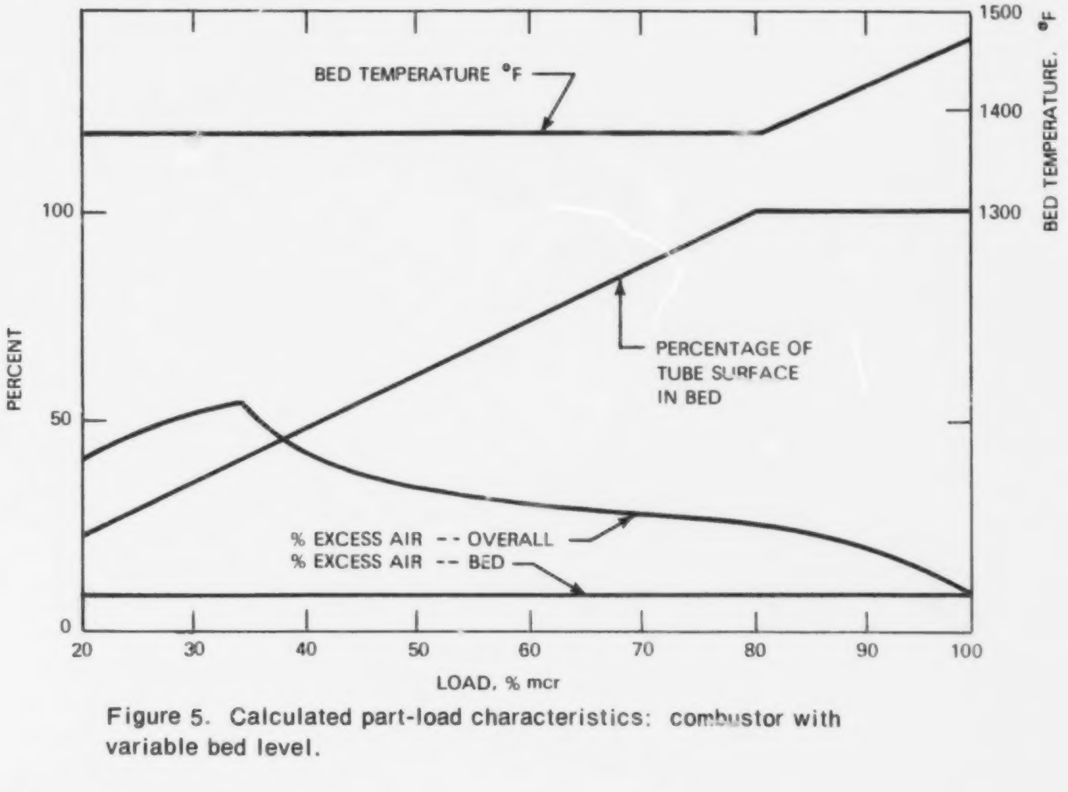


Figure 5. Calculated part-load characteristics: combustor with variable bed level.

5. COMPARATIVE EMISSIONS OF POLLUTANTS DURING COMBUSTION OF NATURAL GAS AND COAL IN FLUIDIZED BEDS

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ABSTRACT

The feasibility of employing a fluidized-bed combustor for the combustion of natural gas is being investigated. Because of the relatively low temperature (1600°F) usable in this method of combustion, NO generation is held to the level dictated by the nitrogen fixation equilibrium, about 60 ppm. Combustion efficiencies in the range 94-99 percent were obtained; however, a combustion efficiency of greater than 99 percent can undoubtedly be achieved by more efficient distribution of natural gas in the fluidized bed.

The reaction of artificially generated SO_2 with limestone during natural gas combustion at 1600°F resulted in SO_2 removals of 95 to 99 percent at a low Ca/S mole ratio of 1.5. These excellent results were probably due to more efficient distribution of the injected SO_2 in the combustor and to the ash-free environment.

In other work, it was noted that when the bed of partially reacted limestone was heated, desorption of SO_2 started at slightly above 1700°F. At 1850°F with 1 vol percent oxygen in the flue gas, SO_2 concentration in the flue gas was 5000 ppm. The reason for this high degree of SO_2 desorption at a relatively low temperature (a temperature of about 2200°F was thought necessary to obtain this effect) may be the presence within the bed of partial reducing conditions at the low excess-air level employed.

Higher than equilibrium concentrations of NO have been observed during fluidized-bed coal combustion. The source of the nitrogen was shown to be the nitrogenous content of the coal itself—argon was substituted for the nitrogen content of the combustion air and no significant effect on NO concentration was observed.

A concomitant reduction in NO concentration with a reduction in SO_2 concentration upon the addition of limestone has been noted. This is probably related largely to the presence of sulfated limestone.

The use of metal oxides (Al_2O_3, Co_3O_4, and ZrO_2) as catalysts for the decomposition of NO was not effective. In fact, when Co_3O_4 was tested, the NO concentration in the flue gas increased.

The results of three independent analytical procedures showed that nitric oxide is the only nitrogen oxide species present in the flue gas in a significant quantity during the fluidized-bed combustion of coal.

INTRODUCTION

Natural gas fuel is increasingly utilized in power generating plants as a means of eliminating the SO_2 emission problem; however, NO concentration in the flue gas is high due to the reaction of oxygen and nitrogen at the elevated temperature of the gas flame. The

NO concentrations observed are in the range 160-1140 ppm, indicating that combustion temperatures are as high as 2000-3000°F. Because of increasing concern about the emission of nitric oxide from electric power generating stations, the feasibility of natural gas combustion in a fluidized-bed combustor has become of interest. Combustion of natural gas in a fluid-bed combustor could be efficiently performed at relatively low temperatures (e.g., 1600°F), and conceivably only the equilibrium NO concentration of <100 ppm would be in the gas.

To test the feasibility of fluid-bed combustion of natural gas as a means of reducing NO emissions, experiments have been performed at Argonne in a 6-in. diameter bench-scale combustor. The results of these experiments are presented here, along with ancillary investigations concerning the behavior of a partially sulfated limestone bed during natural gas combustion. The data obtained with partially sulfated limestone beds suggests answers to such questions as:

1. How does the location of SO_2 generation in the fluid bed affect SO_2 removal?
2. Why is SO_2 released from partially sulfated limestone at temperatures considerably below those expected from equilibrium considerations?

The final section of this paper discusses general experience pertaining to NO formation and removal from the flue gas in fluidized-bed experiments with coal and natural gas flues.

APPARATUS AND MATERIALS

The 6-in. diameter bench-scale combustor is described in detail in a recent annual report on this program.[1] Briefly, as shown in Figure 1, the bench-scale equipment consists of a combustor (about 6 ft long), an air preheater (also 6 ft long, but only 6 in. in diameter), vibratory screw feeders for delivering the coal or limestone additive into an air transport stream for injection into the bed, cyclone separators and a glass fiber final filter to remove entrained solids from the flue gas, and instruments for gas metering and gas analysis.

In the startup procedure for these bench-scale experiments, the fluidizing air was preheated to about 1000°F; the heated air increased the bed temperature to 700-800°F. A bed temperature of about 1200°F is necessary for the ignition and sustained burning of natural gas. For the natural gas combustion experiments, the bed temperature was raised further by burning either coal or propane so that the methane could ignite. Subsequently, natural gas was injected into the fluidized bed at the coal injection point (about 4 in. above the fluidizing-air distributor plate). Upon ignition and sustained burning of the natural gas, the fluid-bed temperature was raised to the operating range of 1600-1850°F.

The natural gas, from a commercial gas line, was supplied by Northern Illinois Gas Co., who also supplied a typical analysis (Table 1) accomplished by gas chromatography. The nitrogen present as elemental nitrogen was added to adjust the heat content value of the natural gas.

The fluid bed for the natural gas combustion experiments was the final bed from a previous coal combustion experiment in which a fine-particle-size limestone had been reacted with SO_2. The calcium and sulfur contents of this bed were 1.54 and 0.95 wt percent, respectively; the remainder of the bed was essentially refractory alumina.

In several experiments in the natural gas series, SO_2 was added to the gas phase. Purity of the SO_2 was >99 percent. The limestone No. 1359 used had an average particle size of 25 μm and contained 97.8 wt percent $CaCO_3$ and 1.3 wt percent $MgCO_3$.

NATURAL-GAS COMBUSTION EXPERIMENTS

NO Level and Combustion Efficiency

The NO concentrations, measured in the flue gas for natural gas combustion experiments at a temperature of 1600°F, varied from 60 to ∿100 ppm at an oxygen concentration in the flue gas of about 3.5 vol percent. (The oxygen concentration in the flue gas reflects the percent excess air in the feed and the combustion efficiency.) With 3.5 vol percent oxygen in the flue gas at 1600°F, the nitrogen fixation equilibrium data would give a NO concentration of 54 ppm. This is in reasonable agreement with the observed levels. This data indicates that significantly lower NO concentrations should be achieved in a fluidized-bed natural gas combustor than in a conventional combustor.

Table 1. COMPOSITION ANALYSIS OF NATURAL GAS

Constituent	Analysis[a] by Gas Chromatography, vol %
CH_4	88.15
C_2H_6	5.38
C_3H_8	1.65
C_4H_{10}	0.43
C_5H_{12}	0.10
CO_2	0.36
He	0.17
O_2	0.05
Miscellaneous	0.07
N_2	3.64
Heat content, Btu/cu ft	1035
Specific gravity	0.6

[a] Northern Illinois Gas Co.

Combustion efficiency of the natural gas at 1600°F was about 99 percent with oxygen concentrations in the flue gas of 3-4 vol percent. More efficient distribution of natural gas in the fluid bed could no doubt increase combustion efficiency above 99 percent. The efficiencies were calculated from CO and hydrocarbon analyses (obtained by gas chromatography) of grab samples of flue gas. The concentrations of CO and hydrocarbon (expressed as CH_x) in the flue gas at 99 percent combustion efficiency averaged about 900 and 250 ppm, respectively. As would be expected, the combustion efficiency was sensitive to oxygen concentration in the flue gas; at 1-2 vol percent oxygen, the combustion efficiency averaged 94% and the CO and CH_x concentrations averaged 6000 and 2600 ppm, respectively. These results are in general agreement with those obtained in a Russian application[2] of fluidized-bed combustion of gas to heating furnaces.

Desorption and Reaction of SO_2 with Limestone

During the course of a natural gas combustion experiment (Figure 2), SO_2 began to desorb from the partially sulfated bed material when the bed temperature was ∿1720°F. Desorption increased rapidly and reached a peak concentration in the flue gas of 2450 ppm at a temperature of 1820°F, at which point the oxygen concentration in the flue gas was 1.3 vol percent. When the oxygen level was increased to 4.4 vol percent, SO_2 concentration in the flue gas decreased to 200 ppm. Of potential interest for elucidating the mechanism of this desorption reaction are the high concentrations of CH_4 and CO at the stage where SO_2 desorption was near its highest value (see Figure 2). The presence of the reducing gas CO might contribute significantly to desorption.

Both combustion temperature and oxygen concentration are variables that could affect the desorption of SO_2. In another experiment, with a constant 1.8 vol percent oxygen concentration in the flue gas, the SO_2 concentration was 2250 ppm at 1840°F and 800 ppm at 1790°F. When the combustion temperature was maintained at 1850°F, Table 2 shows that the SO_2 concentration varied inversely with oxygen concentration in the flue gas over a rather narrow oxygen concentration range. Figure 3 shows the dependence of SO_2 concentration in the flue gas on oxygen concentration. It shows that doubling the oxygen concentration (from 1.0 to 2.0 vol percent) reduces the SO_2 desorption by about half, and that a fourfold increase in oxygen concentration (from 1 to 4 vol percent) decreases the desorption of SO_2 tenfold. Integration of the SO_2-time desorption curve of Figure 2 indicates that approximately 30 percent of the sulfur originally contained in the fluid-bed material was desorbed during the short period of the desorption phase of the experiment.

The relative ease of desulfurization of the bed material is potentially of high significance. It suggests that the regeneration of sulfated limestone and the recovery of sulfur values might be readily achieved in an auxiliary reactor operating as a combustor at slightly higher temperature, and at a low oxygen concentration. The use of highly reducing conditions of regeneration apparently is not necessary.

A series of natural-gas combustion experiments was performed to investigate the effect of the absence of fly ash on the extent of SO_2 removal. Sulfur dioxide was injected into the fluidizing air stream near the preheater at a flow rate sufficient to result in a flue gas concentration of 3000 to 4000 ppm in the absence of additive. Limestone was then injected into the fluid bed, and the extent of SO_2 removal was observed.

Table 2. VARIATION IN SO_2 CONCENTRATION IN FLUE GAS AS A FUNCTION OF OXYGEN CONCENTRATION AT 1850 °F

O_2 in Flue Gas, vol %	SO_2 in Flue Gas, ppm
1.8	2700
1.5	2820
1.0	4280
0.5	4900
1.8	2050

Figure 4 shows the results for an additive injection period of a typical natural gas combustion experiment (NG-3) at 1600°F. The starting SO_2 concentration of 3520 ppm was reduced to about 160 ppm, an SO_2 removal greater than 95 percent. Because of uncertainty in the zero point of the infrared detector for SO_2, the actual SO_2 removal may have been as high as 99 percent. The CaO utilization for limestone No. 1359 (average particle size, ∿25 μm) was 66 percent. Both the extent of SO_2 reduction and the limestone utilization are higher than were obtained during combustion of coal, especially since a rather low Ca/S mole ratio of 1.5 was used. A similar coal combustion experiment at the same Ca/S mole ratio would be expected to result in removal of only 60 percent of the SO_2.

The SO_2 removal by reaction with limestone appeared to depend on where SO_2 is injected into the bed. In two experiments at 1600°F with limestone No. 1359 and Ca/S mole ratios of 1.8 and 2.1, SO_2 was injected directly into the combustor; SO_2 removals from the flue gas were 95 and 92 percent, respectively. The major difference between the sets of experiments discussed in this and the preceding paragraph was the location in the apparatus where SO_2 was injected. For the experiment (discussed above) which resulted in 95 to 99 percent removal of SO_2, the SO_2 was injected into the fluidizing air at the preheater, upstream of the combustor. For the other experiment, which resulted in a lower SO_2 removal, the SO_2 was injected into the combustor near the point where natural gas was injected into the reactor. These results suggest that the reason for a greater degree of SO_2 removal was more efficient mixing and homogenizing of the gas mixture related to the mechanics of injecting the SO_2 into the fluidizing air.

The possibility that added fine particulate material affects SO_2 removal during natural gas combustion was investigated. The high levels of SO_2 removal which were obtained in the natural gas experiments were for cases in which no fly ash was present. Tested for their effect on SO_2 removal were (1) fine alumina (≤44 μm) and (2) fly ash produced in a coal combustion experiment and collected in a cyclone separator. Table 3 lists experimental conditions and results obtained. In this experiment, SO_2 was added at a 1600°F combustion temperature until flue gas concentration was 3840 ppm. At this point, limestone No. 1359 was added at a rate to achieve a Ca/S mole ratio of 1.8, resulting in an SO_2 removal of 95 percent. Fine alumina was then injected into the reactor at a rate of 1.4 lb/hr, approximately three times the usual rate of fly-ash production during a coal combustion experiment. The presence of the fine alumina

Table 3. EFFECT OF ADDITION OF FINE PARTICLES ON SO_2 ADSORPTION BY LIMESTONE IN A NATURAL GAS COMBUSTION EXPERIMENT[a]

Added Solids	Solids, lb/hr	Limestone No. 1359, lb/hr	Ca/S Mole Ratio	SO_2 in Flue Gas, ppm	% SO_2 Removal	% CaO Utiliz.
–	–	0	0	3840	–	–
–	–	0.8	1.8	180	95	53
Fine Al_2O_3	1.4	0.8	1.8	180	95	53
Fly ash	0.9	0.8	1.6[c]	∿400	∿90	∿56
Fly ash	0.9	0.8	1.6[c]	∿400	∿90	∿56
Fly ash	1.7	0.8	1.4[c]	∿620	∿84	∿60

[a] Experiment conditions:

Starting bed	29.1 lb sintered alumina (30 mesh)
Bed height	15 in. static and 24 in. fluidized
Fuel	Natural gas, Northern Illinois Gas Co., 0.82 scfm,[b] plus SO_2 added at 1.7 scfh.[b]
Additive	Limestone No. 1359, 25 μm, 97.8 wt % $CaCO_3$
Superficial gas velocity	2.7 ft/sec
Temperature	1600°F

[b] STP at 70°F, 1 atm.

[c] Fly ash contained 2.4 wt % S.

particles did not affect the reaction of SO_2 with limestone. Fly ash was then injected into the combustor in three separate injection periods: during the initial two periods, at a rate twice that of fly-ash production during coal combustion; and during the third period, at a rate 3.5 times the normal fly-ash production rate during coal combustion. The removal of SO_2 was lowered to 90 percent during the periods at the lower fly-ash injection rate, and to 84 percent during fly-ash injection at the higher rate. In this experiment, the effect of the presence of fly ash on SO_2 removal, although significant, did not completely account for the observed difference between the natural gas experiments and the coal combustion experiments (in which the extent of SO_2 removal was only about 65 percent). However, the effect on SO_2 removal of fly ash produced on site may be quite different (e.g., the actual effect may be greater) from that indicated by these experimental results.

Conclusions–Experiments with Natural Gas Fuel

1. Fluidized-bed combustion of natural gas at 1600°F results in NO concentrations in the flue gas that are essentially those expected from the nitrogen fixation equilibrium.

2. The extreme simplicity of applying the fluidized-bed concept to combustion of natural gas in a fluid bed is noteworthy. Because ash is absent and because the fuel is easily distributed throughout the fluidized bed, scale-up would be simpler than for coal. Only small-scale experimentation would be needed to optimize the operating conditions for high combustion efficiency. Hence, if a market exists for this concept, it would represent a means for the early demonstration of fluidized-bed combustion on a prototype scale.

3. The reaction of injected SO_2 with limestone during natural gas combustion resulted in a high degree of SO_2 removal from the flue gas. Most probably, the 95 percent or higher SO_2 removal is largely due to better SO_2 distribution in the combustor and to the absence of fly ash in this method of combustion.

4. By operation of a natural gas combustor at 1850°F with a low oxygen concentration (1 to 2 vol percent) in the flue gas, SO_2 might be desorbed from sulfated limestone. This mode of combustion is effective in desorbing SO_2 at a temperature several hundred degrees below that at which desorption is expected, and is thought to be a result of partial reducing conditions in the bed.

NITROGEN OXIDES IN THE FLUIDIZED-BED COMBUSTION OF COAL

Source of Nitrogen

Nitrogen oxides, principally NO, are formed during the combustion of fossil fuels and are important contributors to air pollution. Although the quantities of NO observed in the flue gas from high-temperature conventional combustors can be accounted for by the equilibrium of the nitrogen fixation reaction, this is not the case for fluidized-bed coal combustors, in which the concentrations of NO have been observed to be far in excess of those expected on the basis of the equilibrium. At 1600°F, a common temperature for fluid-bed combustion, the equilibrium concentration of NO should vary from 50-200 ppm, depending on the oxygen concentration. The oxygen concentration, in turn, would depend on the level of excess air employed. However, in actual fluid-bed coal combustion, NO levels of 400-800 ppm in the flue gas have been measured.

One possible explanation of these high NO concentrations is that the temperature of the combusting particles might be higher (e.g., 1800-2100°F) than the measured temperature, 1600°F. Another possible explanation for higher than equilibrium NO concentrations is that the nitrogenous content of the coal (1-1.5 wt percent in U.S. coals) reacts with oxygen in the fluidizing air to form the excess NO. That coal is indeed the source of nitrogen was demonstrated by substituting an inert component, argon, for the nitrogen component of the fluidizing gas. Several fluid-bed experiments were performed substituting argon, and it was found that the change from nitrogen to argon did not affect the NO level significantly, showing clearly that most of the NO originated from the nitrogenous content of the coal. In one experiment, the initial NO concentration of 580 ppm did not vary during the argon substitution by more than ±20 ppm. In another, the initial NO concentration of 400 ppm actually increased to 450 ppm when argon was substituted for nitrogen in the fluidizing gas.

This conclusion was substantiated by the results of the previously described natural gas experiments. In these experiments, in which the only source of nitrogen was the fluidizing air, the observed NO concentration in the flue gas, <100 ppm, was that expected from the nitrogen fixation equilibrium.

Removal of NO by Limestone Additive

Another interesting finding is that the NO concentration was reduced concomitantly with the SO_2 removal when limestone was added. The original NO concentration in the flue gas was lowered by as much as 40 percent. That the removal of NO was influenced by the addition of limestone and therefore possibly related to the reduction in SO_2 concentration was indicated by a rise in the NO concentration when the limestone addition was terminated.

The behavior of the NO concentration during the addition of limestone to a fluid-bed combustor was observed in a series of coal combustion experiments. The most significant reduction in NO concentration (to 200-300 ppm from an initial level of about 600 ppm) was achieved when the initial fluid bed was composed entirely of fresh limestone. An interesting observation was that NO removal did not occur immediately after the addition of limestone but only after SO_2 removal had occurred. Possibly, the agent responsible for NO removal is the sulfate formed by the reaction between SO_2 and limestone.

Information has also been obtained showing that the extent of NO reduction is inversely proportional to the Ca/S mole ratio. Figure 5, in which the reduction of NO emission is plotted against the Ca/S mole ratio, shows that as the Ca/S mole ratio increased, the extent of NO removal decreased. A mechanism for the effect is not yet known.

An analysis of a series of fluid-bed experiments showed that the degree of reduction in NO concentration was also related to the velocity of the fluidizing gas. These results (Figure 6) show that NO removal in the flue gas is inversely proportional to superficial gas velocity, suggesting that gas residence time in the bed is a factor in NO decomposition.

A proposed mechanism for the removal of NO concomitantly with the removal of SO_2 upon the addition of limestone in a fluid-bed combustor is the equilibrium reaction $NO_2 + SO_2 \rightleftarrows SO_3 + NO$, which has been studied by British workers.[3] If this reaction occurs, a decrease in SO_2 concentration could result in a corresponding decrease in NO concentration and an increase in NO_2 concentration. This hypothesis was tested by determining the NO_2 content of the flue gas during a fluid-bed combustion experiment. Three different methods were used to assay the NO_2 content of the flue gas:

1. The phenoldisulphonic acid method.
2. Mass spectrometric analysis.
3. The Mast NO_2 analyzer.

Analyses were performed on samples of flue gas taken both before and after the addition of limestone in a coal combustion experiment. The results from all three methods indicated that essentially no NO_2 was present in the flue gas samples taken. For example, mass spectrometric analysis indicated that the NO/NO_2 ratio was at least 50/1 and probably much higher. On the other hand, the Mast in-line NO_2 analyzer consistently indicated no NO_2 or at the most concentrations only in the ppm range. Of the three methods, the phenoldisulfonic acid method seemed the least precise; by difference ($NO_x - NO = NO_2$), both negative and positive values were obtained for the NO_2 concentration.

Investigation of Catalytic Decomposition

Catalytic decomposition was considered as a means of controlling NO emission. The literature in the field of catalytic control of nitrogen oxides[4, 5] indicates that low reaction rates, instability of the catalyst at fluid-bed temperatures, and catalyst poisoning by sulfur oxides are among the factors mitigating against on-site use of particulate catalysts. Nevertheless, the attractiveness of this approach for lowering NO emission, the fact that these catalysts had not been previously evaluated in a fluidized bed, and the simplicity of the testing procedures led to the performance of scoping tests. The literature indicated that oxides of aluminum, cobalt, and zirconium appeared to be the most effective of the decomposition catalysts.

Experiments were conducted in the bench-scale fluid-bed combustor by injecting solid metal oxides into the fluidized-bed combustor at the same location at which the coal and limestone additive were injected. In the first experiment, the solid catalysts used were (in order of their use) aluminum oxide (Al_2O_3), zirconium oxide (ZrO_2), and cobalt oxide (Co_3O_4, cobaltous-cobaltic oxide). Except for zirconium oxide, which contained 1-2 percent hafnium, these materials were reagent grade with purities greater than 99.9 percent.

Injection of Al_2O_3 or ZrO_2 into the fluidized bed at rates of several pounds per hour did not change the NO concentration in the flue gas. Injection of each material was continued for about 1 hour.

The injection of Co_3O_4 (cobaltous-cobaltic oxide) at a rate of about 4 lb/hr caused the NO concentration in the flue gas to increase rather than decrease (the latter would be expected if NO were catalytically decomposed). The NO concentration increased from about 700 ppm before injection to about 1400 ppm during injection. When Co_3O_4 injection was stopped, the NO concentration in the flue gas dropped back nearly to its original level. Upon reinstitution of catalyst injection, NO concentration in the flue gas immediately increased.

In a second experiment with Co_3O_4, NO and SO_2 concentrations in the flue gas prior to catalyst injection were 600 ppm and 1000 ppm, respectively. When Co_3O_4 was first injected at a rate of about 4.5 lb/hr, the NO concentration increased to about 1000 ppm, an increase of about 400 ppm. Co_3O_4 might act as a catalyst for the oxidation of nitrogen compounds in coal to NO, similarly to the catalytic oxidation of ammonia. In all experiments, the addition of metal oxide catalysts had no discernible effect on SO_2 concentration in the flue gas.

Conclusions—Nitric Oxide Behavior in Coal Combustion Experiments

1. The concentrations of NO observed in fluidized-bed combustion of coal were higher than those predicted on the basis of the equilibrium. A major source of nitrogen was the nitrogenous content of the coal itself.

2. Upon the addition of limestone, nitric oxide concentration in the flue gas is lowered

concomitantly with SO_2 concentration. This reduction in NO level appears to be related to sulfation of limestone.

3. NO is the predominant species if not the only nitrogen oxide species in the flue gas.

4. The use of a metal oxide (Al_2O_3, Co_3O_4, or ZrO_2) as a catalyst for the decomposition of NO was not effective. In fact, the NO level in the flue gas was increased rather than decreased in the presence of Co_3O_4.

5. Evidence was obtained that the equilibrium reaction $NO_2 + SO_2 = NO + SO_3$ was not involved in the reduction of NO concentration.

6. For a Tymochtee dolomite, the extent of reduction of NO concentration during limestone addition was shown to be inversely related to the Ca/S mole ratio; for limestone No. 1359, it was inversely related to the superficial fluidizing gas velocity.

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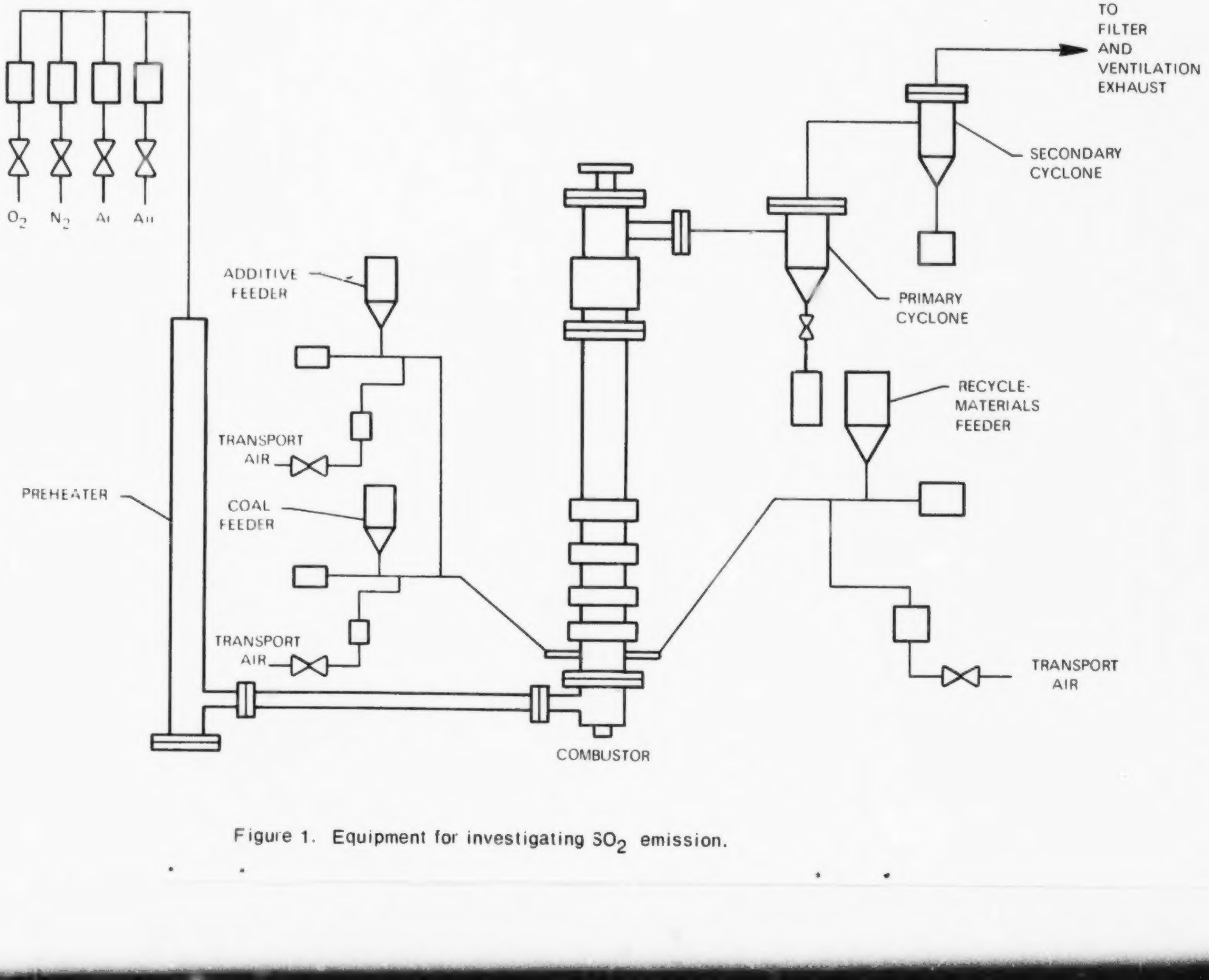
O_2
N_2
Ar
Air
TO FILTER AND VENTILATION EXHAUST
SECONDARY CYCLONE
PRIMARY CYCLONE
ADDITIVE FEEDER
TRANSPORT AIR
COAL FEEDER
TRANSPORT AIR
PREHEATER
RECYCLE-MATERIALS FEEDER
TRANSPORT AIR
COMBUSTOR

Figure 1. Equipment for investigating SO_2 emission.

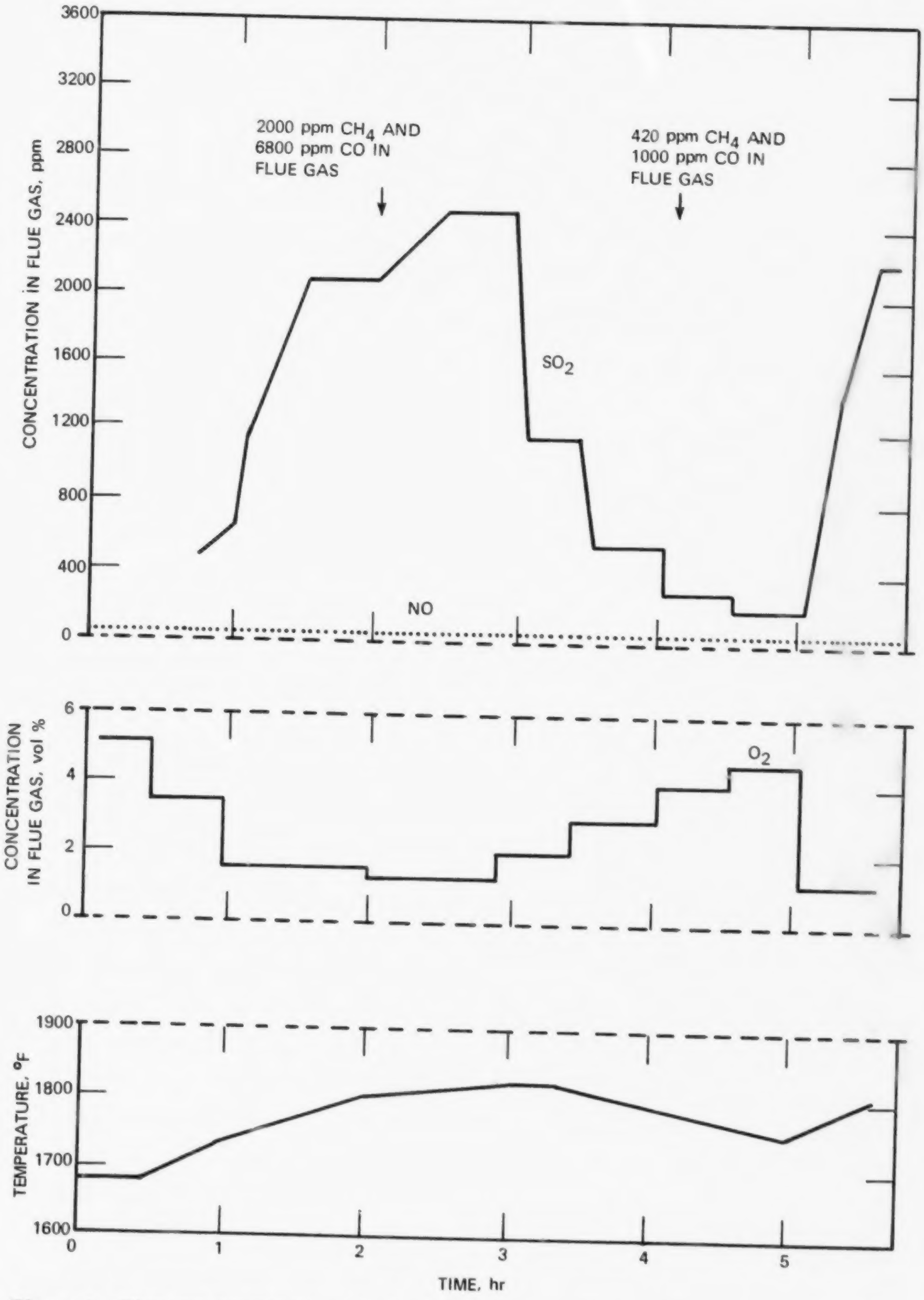

Figure 2. Flue gas composition (dry basis) and combustion temperature, experiment NG-2.

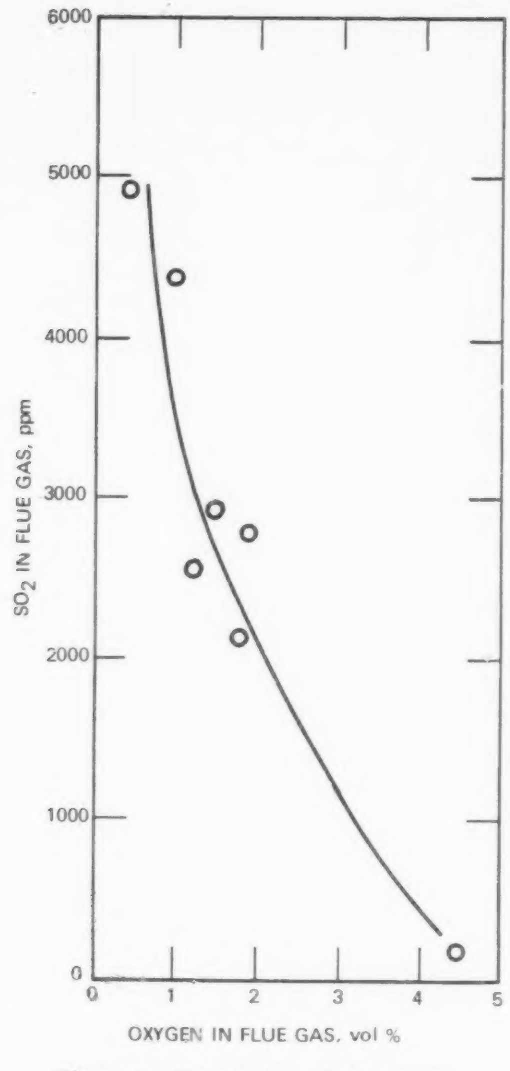

Figure 3. Desorption of SO_2 during natural gas combustion as a function of oxygen concentration at 1820 - 1850 °F.

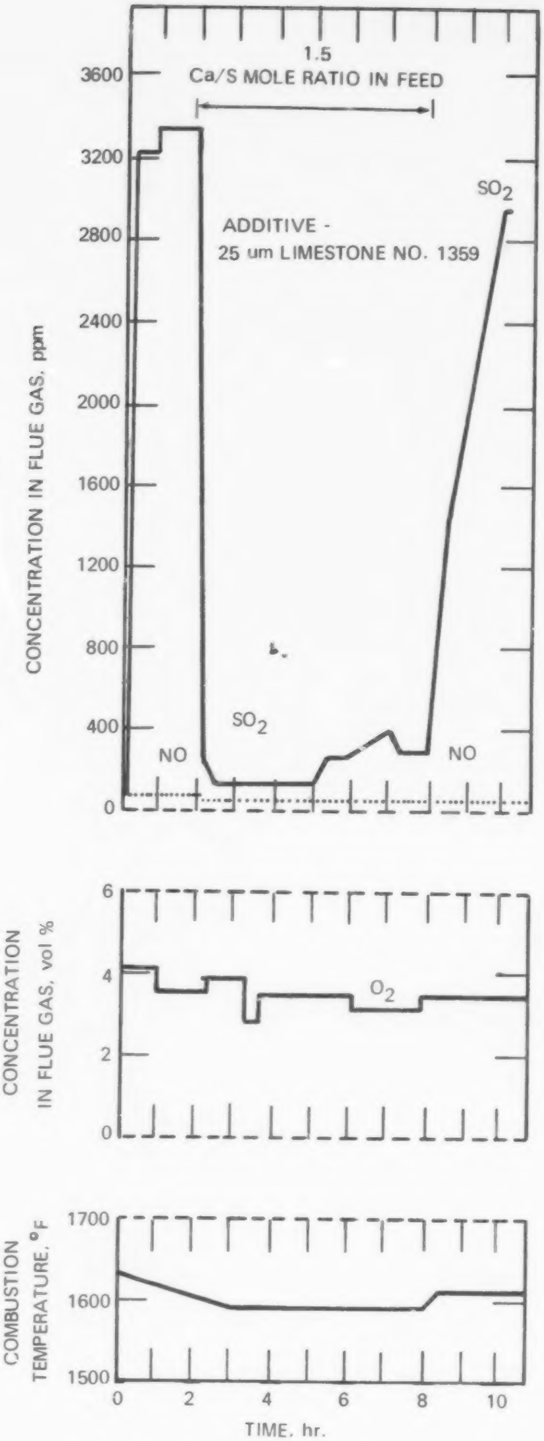

Figure 4. Operating conditions and flue gas composition, experiment NG-3, part 2.

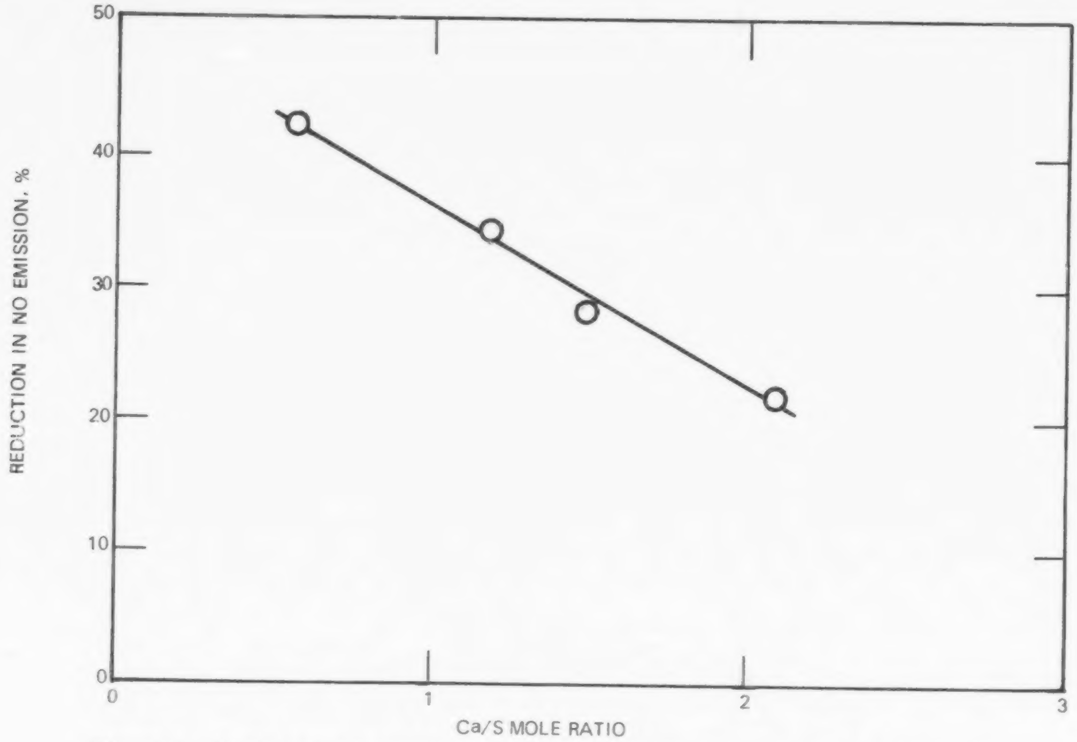

Figure 5. Nitric oxide reduction with -325 mesh (<44 μm) Tymochtee dolomite.

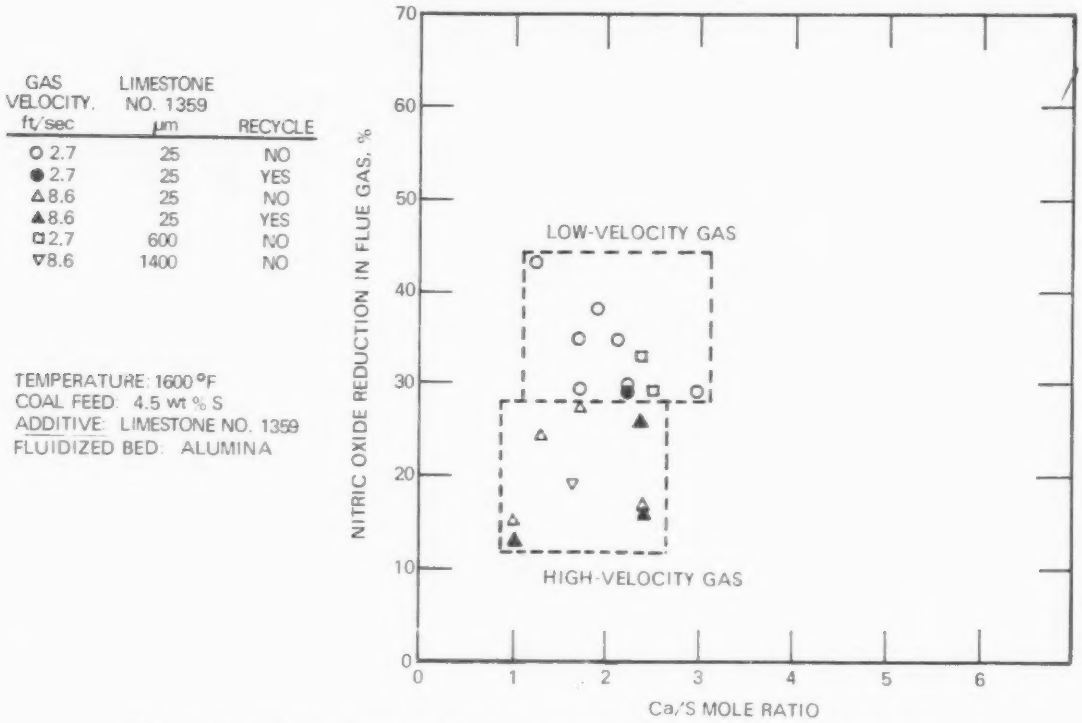

Figure 6. Effect of gas velocity on nitric oxide reduction in flue gas.

6. IGNIFLUID CONTRIBUTION TO AIR POLLUTION CONTROL

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INTRODUCTION

The Ignifluid combustion process, characterized by the fact that the total amount of ash retained comes out as clinker, is operational on an industrial scale.

Boilers associated with this kind of furnace are strictly conventional; those at Casablanca Power Station in Morocco are typical (Figure 1).

One of the main advantages of the Ignifluid process is that it makes it possible to burn coals which have a wide variety of characteristics; e.g., volatile matter, ash, and grain size.

Table 1 gives the characteristics of the Ignifluid units we shall refer to in this paper; Table 2 gives the grades of coal (from anthracite to bituminous) fired in them.

The operating results obtained in all of these different industrial installations can, therefore, very often be considered as having a general character.

Table 1. CHARACTERISTICS OF IGNIFLUID PLANTS

Location	Steam Output M.C.R., lb/hr	Pressure, lb/sq in.	Superheat Temperature, °F	Operating Hrs October 1970
La Taupe	77,000	840	915	5,000
Casablanca	254,000	1,300	1,000	15,000
Solvay	110,000	290	570	8,500
La Rochette	110,000	670	825	53,000

Table 2. COAL USED

Location	Type of Coal	Characteristics of Coal			
		G.C.V., Dry Basis, Btu/lb	V.M., %	Ash, %	Sulphur, %
La Taupe	Middlings from Auvergne	11,000	10.0	22	1.0
Casablanca	Anthracite from Djerada	11,200	6.5	23	2.0
Solvay	Bituminous from Lorraine	13,300	35.0	7	0.7
La Rochette	Anthracite from La Mure	11,200	5.5	21	0.5

It is not the purpose of this paper to deal with the Ignifluid process from the combustion point of view (e.g., its efficiency, flexibility in operation, and availability). These subjects have already been discussed: by Mr. (A.A.) Godel at the first International meeting 2 years ago, as well as in more recent publications.

It is generally agreed that the fluidized-bed combustion process may be an efficient means of overcoming atmospheric pollution. It is interesting to examine the position of the Ignifluid system with regard to this important problem.

GOAL–TO LIMIT FLY ASH

Historically speaking, in the course of the 15 years devoted to the development of the Ignifluid process, our efforts from this point of view are aimed solely at limiting the solid fly ash at the stack. The reason is that French coals, like most European coals, have the great advantage of containing very little sulphur. On the other hand practically all the Ignifluid furnaces have been installed in France or abroad for firing coal with a high ash content.

For units with a steam output of 150,000 lb/hr or less, mechanical dust collecting has proved efficient enough. This is mentioned only as a reminder.

The interesting results to be considered are, therefore, those obtained with the electrostatic precipitator at the 60-MW Power Station at Casablanca in Morocco, fitted with Ignifluid furnaces burning anthracite fines with more than 20 percent ash.

Various measurements which have been carried out make it certain that the efficiency guarantee (240 mg/Nm3 or 0.105 grains/std cu ft) is easily reached (160 gr/Nm3 or 0.070 grains/std cu ft).

At the present time, very exhaustive tests are being carried out in Casablanca: we hope they will provide us with more useful data.

These results are mainly due to the intrinsic characteristics of the Ignifluid grate and particularly:

1. The large-sizing particles of the fly ash–compared with the fly ash from pulverized coal furnaces, considerably facilitates the work of the precipitators by natural settling. The dust content at the outlet of the precipitator is practically independent of the dust content in the gas at the inlet (for high-ash coals) because the fly ash increase, according to the boiler load, occurs through the addition of coarse grains. As an example, the above-mentioned efficiency is obtained with a very high dust content before the precipitator, of 80 g/Nm3 or 35 grains/std cu ft.

2. The high carbon content in the fly ash is also a favourable factor. It may seem paradoxicall that a high unburnt carbon content in the fly ash (70 percent at Casablanca where the coal fired is anthracite) should be considered as an asset for the combustion furnace from which it comes. The reason is that all fly ash is refired, and that all the ash is discharged in the form of slag with a low carbon content, and therefore with high efficiency.

Other factors, irrevelant to the Ignifluid process, have a good effect:

1. The S content in the coal at Casablanca is higher than in French and European coals. It amounts to approximately 2 percent: 0.82 percent as pyrites; 1.06 percent as organic matter; and 0.12 percent as sulphate. The general belief that precipitator efficiency increases with the sulphur content is at least in favour of this "impurity." From this point of view, American bituminous coals, with their high sulphur content, should not offer any difficulty for dust removal.

2. The gas temperature in the precipitators, which is approximately 150°C (300°F), is apparently fairly close to the optimum value.

3. Finally, other factors (e.g., steam and alkali content) are doubtless to be taken into consideration.

SO_2/NO_x INTEREST IS RECENT

As mentioned above, only recently has our attention been drawn to the problems of atmospheric pollution due to release of sulphur dioxide (SO_2); and, on another scale, to nitrogen oxides (NO_x).

Moreover, the important project we have been studying for an American company, which Mr. (R.H.) Demmy will tell us about later (in Session No. IV), is based on the use of refuse bank coal with a high ash content (40 percent) and only 1 percent of sulphur.

However, the anxiety expressed throughout the world on the consequences of atmospheric pollution by SO_2, and the interest shown in fluidized-bed combustion methods, have led us not only to examine existing possibilities of the Ignifluid system as it is operated in industrial installations, but also to consider certain possible alterations or additions to make them suitable for more efficient prevention of atmospheric pollution.

The measurements, the results of which are given below, were therefore taken during industrial operation, and not by means of laboratory tests.

The different tests (Table 3) were carried out at La Taupe, Solvay, and Casablanca, in three Ignifluid furnaces, as well as in older boilers equipped with various furnaces: a chain grate stoker at La Taupe, a cyclone furnace at Solvay, and a PF slag tap furnace at Casablanca. Coals fired are those given in Table 2.

The results should be considered comparative, rather than absolute, especially since the S content is generally low.

NO_x Formation

The most interesting result, systematically revealed during the various tests, concerns the nitrogen oxides NO and NO_2 contained in the flue gases of Ignifluid furnaces.

As shown in Table 4, the discharge of nitrogen oxides is two or three times lower than that measured in all the other combustion processes.

These results corroborate the observations that have been made in many existing installations, mainly in the U.S.A., and show that the formation of nitrogen oxides is a function of combustion temperature, excess air, and gas velocities.

Table 3. CHARACTERISTICS OF OTHER PLANTS

Location	Type of Combustion	Steam Output, lb/hr	Pressure, lb/sq in.	Superheat Temperature, °F
La Taupe	Chain grate stoker	77,000	840	915
Casablanca	PF slag tap furnace	190,000	700	860
Solvay	Cyclone furnace	220,000	2,400	1,000

Table 4. NO_x MEASUREMENT IN FLUE GAS

Location	Type of Combustion	Heat Release, 10^6 Btu	NO_x in Flue Gas, ppm	Ratio[a] $NO_x/10^6$ Btu
La Taupe	Ignifluid	90	163	2.68
	Chain grate stoker	84	470	8.45
Casablanca	Ignifluid	237	735	10.70
	PF slag tap furnace	202	940	17.80
Solvay	Ignifluid	107	470	7.55
	Cyclone furnace	245	1720	18.50

[a] NO_x in cuff

The values of these three parameters in Ignifluid combustion combine to reduce the formation of nitrogen oxides to a minimum:

1. Combustion in an Ignifluid bed is completed in two stages –

   a. Combustion in a reducing atmosphere in the Ignifluid bed itself, where a moderate temperature, slightly higher than the mean ash sintering temperature, prevails.

   b. Combustion of the CO gas from the Ignifluid furnace in the area of the secondary air inlet (one or two rows).

   It is moreover well known that the two-stage combustion is an effective way of preventing nitrogen oxides from being formed. It is an idea which has already been applied to the combustion of fuel oil, in particular in the Los Angeles District (B.W. Co. patent No. 821, 214).

2. Combustion in the Ignifluid system takes place with a small amount of excess air because, in the final stage, it consists of burning a gas (CO), which is easier than burning a solid fuel. From this point of view, the Ignifluid system can readily bear comparison with the most efficient furnaces; e.g., the cyclone furnace. This low volume of excess air could be reduced still more if we added certain alterations, to be described later.

3. The long residence time the fuel particles spend in the bed is obviously an essential characteristic of the Ignifluid process.

Finally, without wishing to draw too quick conclusions, it seems feasible to think that the nitrogen oxides content is an increasing function of the heat release, as witnessed by the curves of Figure 2. This phenomenon, already observed, seems to be explained by the fact that the peak temperature in the combustion chamber increases with the size of the units.

SO_2 Release

With regard to the release of SO_2, measurements carried out as in the previous case, with Draeger tubes, show that the result is appreciably the same whatever the type of furnace (except for stokers).

The ratio $\frac{SO_2}{S \times CO_2}$, in which SO_2 is expressed in ppm, S (sulphur in the coal) in percent, and CO_2 in percent, is situated between 50 and 60 (Table 5).

Table 5. MEASUREMENT OF SULPHUR IN FLUE GASES WITHOUT LIME

Location	Type of Combustion	S in the Coal, %	S Retained in Clinker, %	SO_2 in Flue Gas, ppm	Ratio[a] $SO_2/S \cdot CO_2$
La Taupe	Ignifluid	1.00	8.0	635	55.0
		0.55	6.0	400	52.0
	Chain grate stoker	1.00	11.0	890	70.0
Casablanca	Ignifluid	2.00	5.2	425	50.0
	PF slag tap furnace	2.00	2.7	1120	57.0
Solvay	Ignifluid	0.7	2.0	1500	52.5
	Cyclone furnace	0.7	0.2	650	50.0
La Rochette	Ignifluid	0.5	13.0	320	43.0

[a] SO_2 in ppm; CO_2 in %; and S in the coal in %

This observation really shows nothing unusual, taking into account the furnace operating conditions.

It occurred to us to use additives (e.g., calcium oxides or magnesium oxides), projected in the Ignifluid bed in a granulated condition.

A comparatively short test performed in an installation (La Taupe), although giving restricted results, led us to expect some possibilities of development.

These first results (Table 6) show that sulphur retention increases proportionately with the content of lime injected, but it is however still very low, compared with the total amount of sulphur.

These restricted results may be explained by higher temperatures at the rear of the grate (about 1350-1400°C) where an oxidizing atmosphere prevails. With these temperatures, the sulphates created inside the bed decompose releasing SO_2, but the lime remains in the clinker slags.

VARIOUS SO_2 POSSIBILITIES

However, if SO_2 retention must be considered, as is the case with a large number of American bituminous coals, various possibilities seem to be available for the Ignifluid system, in which chemical reactions occur either in the reducing zone or in the oxidizing zone.

Bearing in mind Professor (A.M.) Squires' views on the subject, we believe it would be

Table 6. SULPHUR RETAINED IN CLINKER WITH LIME[a]

Quantity of Lime		S Retained in Clinker, %
Without lime		6
With lime	Twice stoichiometric quantity	8
	Threefold stoichiometric quantity	11

[a] Test at La Taupe

advisable to perform the desulphurization in a reducing atmosphere, where a high conversion of CaO additives to CaS can be achieved.

By recycling flue gases taken at the economiser outlet and reinjected in the last compartments of the grate, an entirely reducing area can be maintained in the lower part of the furnace below the secondary air injection nozzles (Figure 3) at a temperature slightly higher than the sintering temperature (1050 to 1100°C or 1930 to 2000° F).

The recycled flue gas can also be used with primary air to regulate the bed temperature while maintaining the minimum gas flow rates for fluidization.

As another consequence of CaO addition in the bed, a corresponding fluxing action will result in the formation of suitable clinkers.

However, in the complex fluidized bed with heterogeneous conditions, it is difficult to predict the extent to which sulphur will be fixed as sulphide or, perhaps, as sulphate until experimental data is available.

During tests to be performed, parameters other than recycling factors are to be taken into consideration; e.g., the additive grain sizing and injection points (undergrate, that is, in primary air with coal).

Another approach to the SO_2 retention problem is a process described by Mr. Godel during the first International Conference in November 1968, by application of the U.S. patent No. 3,431,892.

The figure (Figure 4) presented by Mr. Godel was comparatively complex; the installation involved many superposed beds. The present arrangement (Figure 5) is limited to a single secondary bed, made of desulphurizing additives in a finely granulated state.

The simpler arrangement is an interesting combination of the Ignifluid process with slagging bed at high temperature and the fluidized combustion system at a lower temperature; this one is controlled by the immersion of boiler tubes in the fluidized bed.

The main advantage of this association consists of the possibility of reinjecting fly ash, consisting of sulphur-free ashes and unburnt particles, into the slagging bed from which all the ashes are finally eliminated in the form of clinkers.

The secondary bed can work in either atmosphere: reducing or oxidizing. Because of the contrast between kinetics for formation of CaS and $CaSO_4$, it appears to be better to operate with a reducing bed where a high conversion of CaO to CaS can be achieved.

According to this program, it is possible to make a valid test, by erecting (along with an Ignifluid conventional boiler) a secondary bed with only 5 to 10 percent of the flue gases, coming out of the boiler used for desulphurization. The desulphurized flue gases, containing a large quantity of CO, would be burnt above the bed (Figure 6) and readmitted, after having been cooled, above the level of the secondary air inlets.

As a conclusion, we may say that the Ignifluid grate presents features that can be used to efficiently control dust and nitrogen oxides emissions.

To overcome the problem of the retention of sulphur in solid form, many tests of various kinds are required.

Although such investigation is not justified in France, we would be willing to draw up a test program, and then carry it out in collaboration with the organisations concerned in one of their Ingifluid plants, on behalf of any other countries where this need occurs.

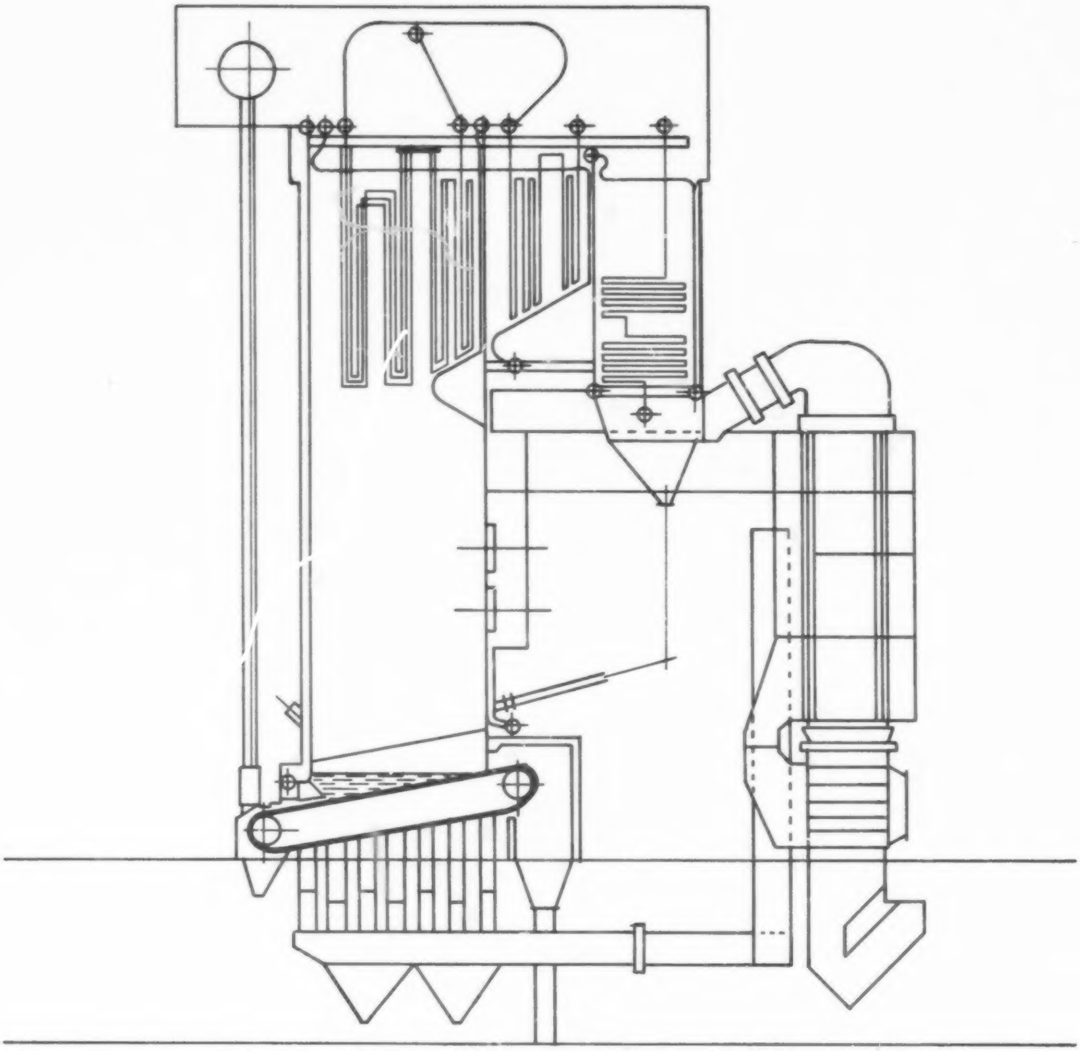

Figure 1. Roches Noires power station at Casablanca.

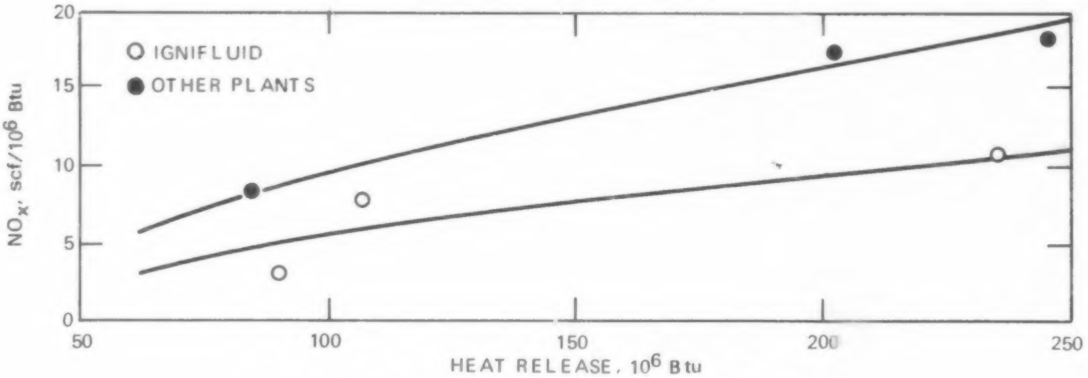

Figure 2. Nitrogen oxides content versus heat release.

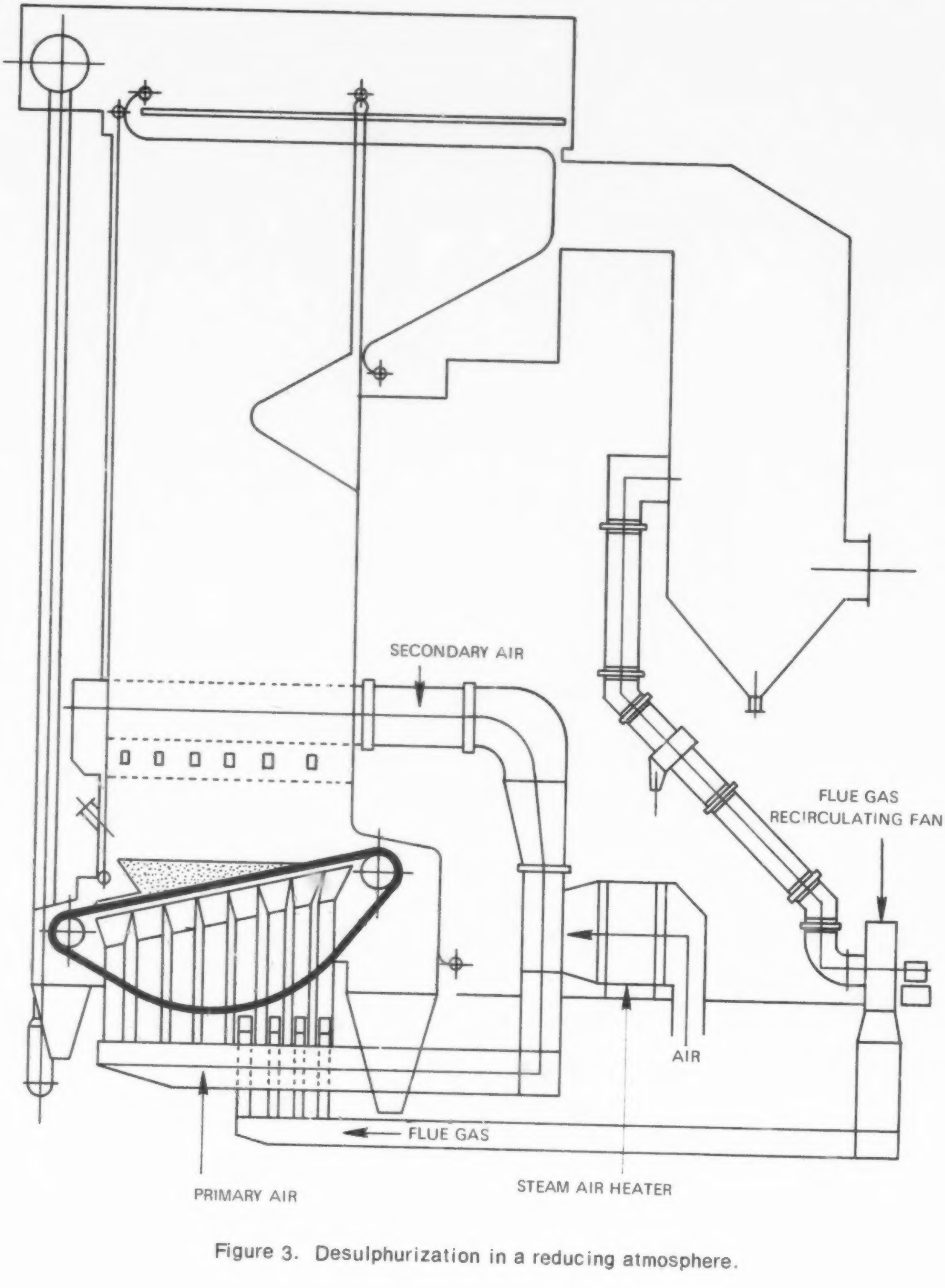
SECONDARY AIR
FLUE GAS RECIRCULATING FAN
AIR
FLUE GAS
PRIMARY AIR
STEAM AIR HEATER

Figure 3. Desulphurization in a reducing atmosphere.

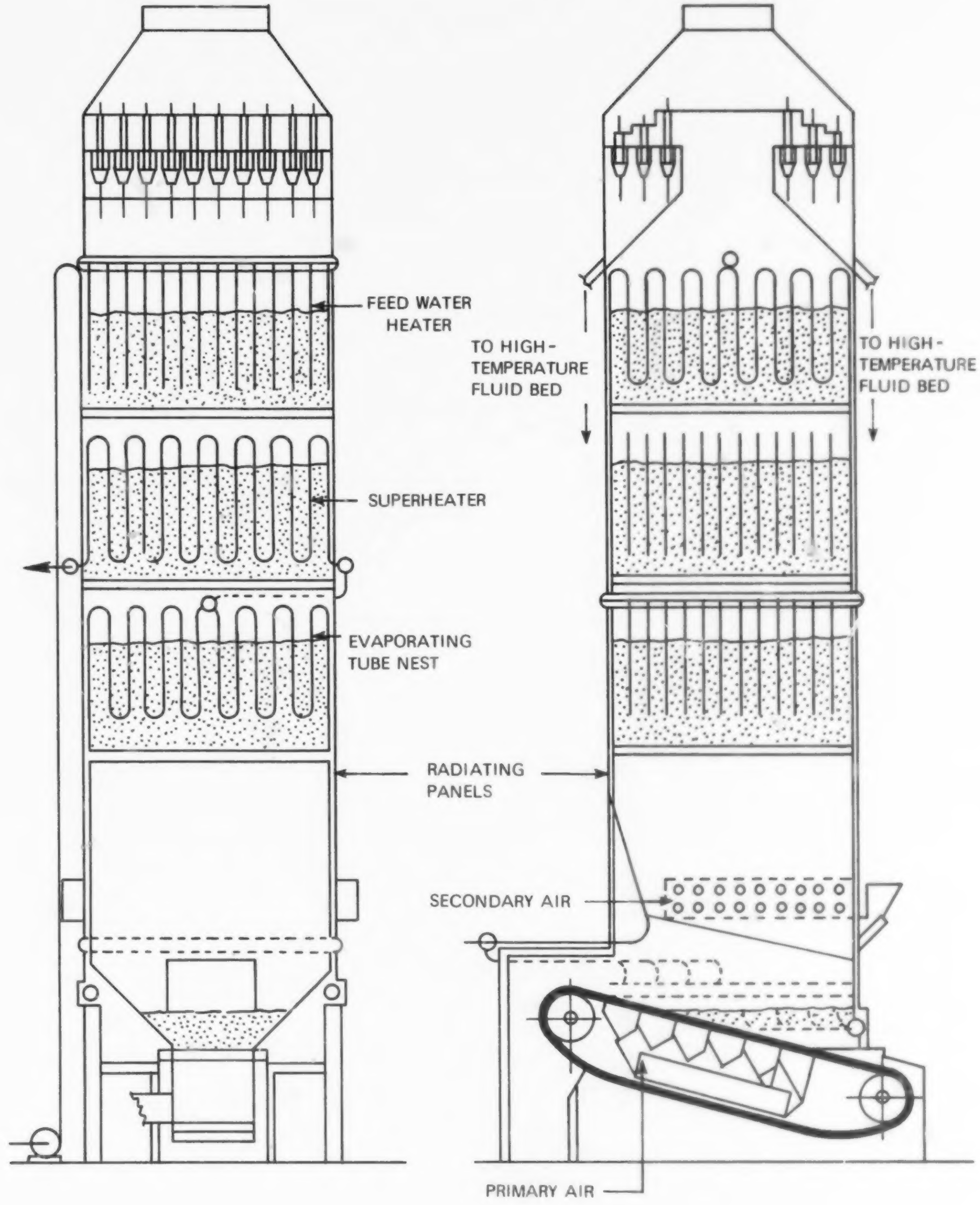
FEED WATER HEATER
TO HIGH-TEMPERATURE FLUID BED
TO HIGH-TEMPERATURE FLUID BED
SUPERHEATER
EVAPORATING TUBE NEST
RADIATING PANELS
SECONDARY AIR
PRIMARY AIR

Figure 4. Former multi-superposed-bed arrangement.

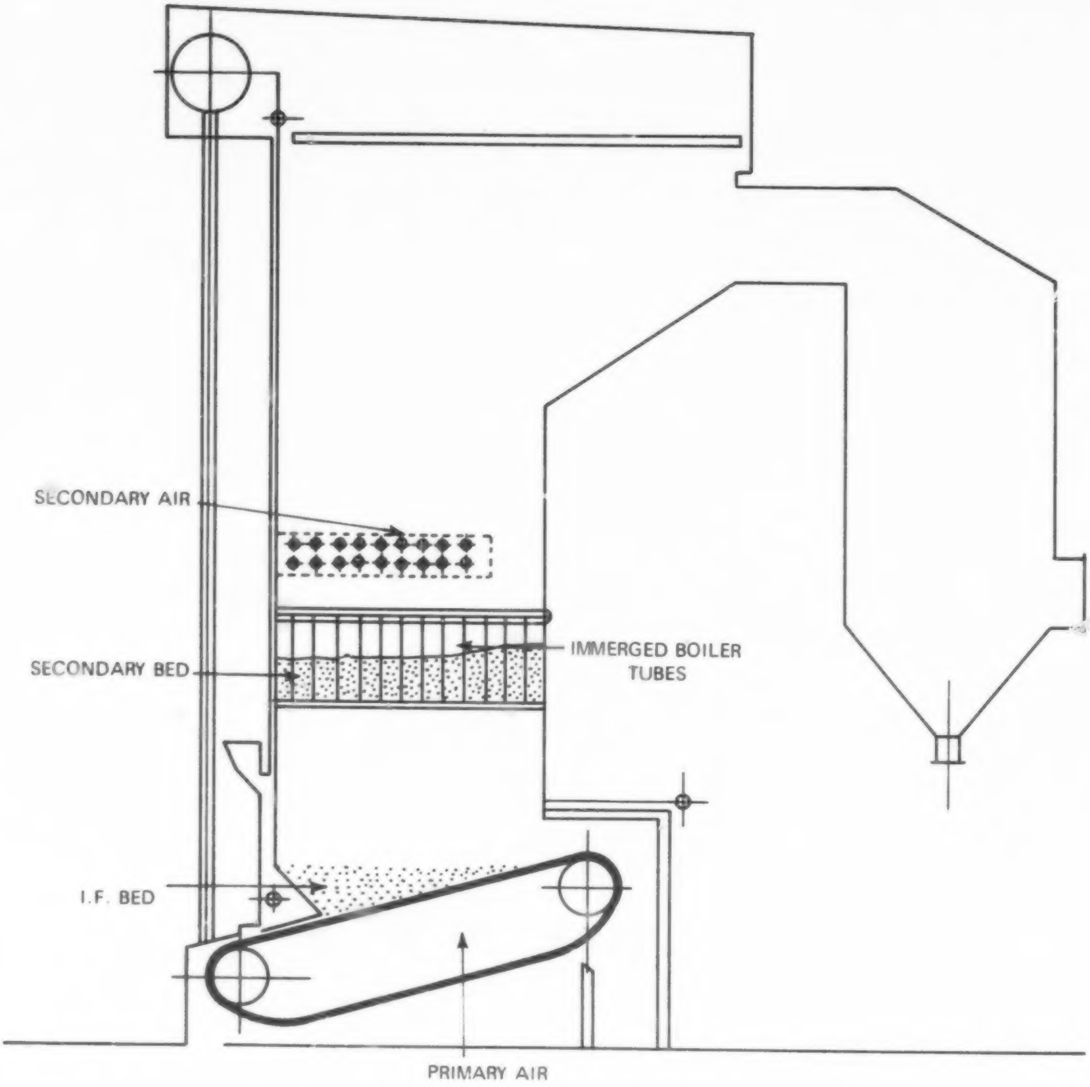
SECONDARY AIR
IMMERGED BOILER TUBES
SECONDARY BED
I.F. BED
PRIMARY AIR

Figure 5. Present single-secondary-bed arrangement.

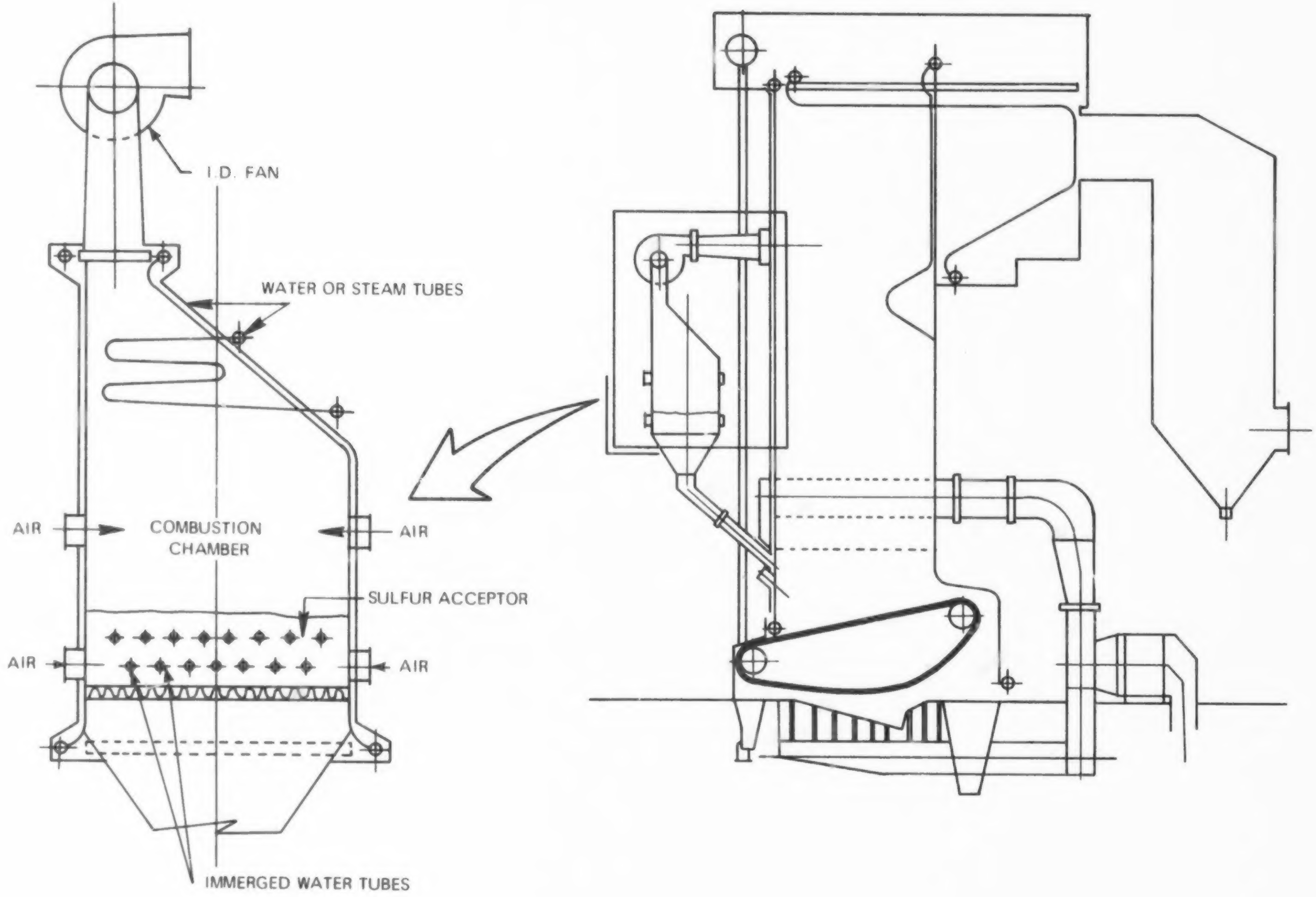
I.D. FAN
WATER OR STEAM TUBES
AIR
COMBUSTION CHAMBER
AIR
SULFUR ACCEPTOR
AIR
AIR
IMMERGED WATER TUBES

Figure 6. Ignifluid conventional boiler and secondary bed test arrangement.

SESSION II:

Control of Combustion Pollutants

SESSION CHAIRMAN:

Mr. Alvin Skopp, Esso Research and Engineering

1. THE RETENTION OF SULPHUR BY LIMESTONE IN A PILOT-SCALE FLUID-BED COMBUSTOR

D. C. DAVIDSON AND A. W. SMALE

National Coal Board, England

INTRODUCTION

The advantages of burning coal in fluidised beds have been widely publicised[1] [2] and much experimental activity has been reported. The fluid bed is an excellent gas/solids contactor and is, therefore, not only an ideal solid fuel combustion system, but also offers attractive possibilities for removing sulphur oxides from combustion gases.

It has been stated[3] that limestone or dolomite addition is the simplest means of removing sulphur oxide from flue gases in existing boilers, both solid fuel and oil fired, but that this simplicity is unfortunately accompanied by inefficiency due principally to very limited residence time in what has been claimed[4] as the effective temperature range (950 - 1230°C; 1770 - 2250°F). This problem is largely overcome by fluidised-bed combustion which is sensibly isothermal. However, the typical operating range of 800 - 900°C (1472 - 1652°F) is, according to Reid's predictions,[4] too low for rapid reaction between limestone and SO_2.

This paper discusses experimental work carried out at the Coal Research Establishment of the National Coal Board; some of this has already been reported[1] in outline. It refers to a current research programme sponsored by the National Air Pollution Control Administration (NAPCA), whose permission to quote results from this work is gratefully acknowledged.

EXPERIMENTAL

The combustor used for the work is shown in outline in Figure 1; a more detailed description of its function is given by McLaren and Williams.[1] The bed is 6 inches in diameter; bed depth is normally controlled at 2 feet. It was possible to recycle fines from a primary cyclone if desired.

The experimental programme has included six coals to date, three from the U. K. and three from the U. S. A. A description of these fuels as fed to the combustor is given in Table 1. In all but one case, the same limestone (U.K.) was used and in all cases the feedstock, both coal and limestone, was nominally -1/16 inch. However, there were differences in the size analyses of the various batches of limestone; these are shown in Table 2. In one series of tests with Peabody No. 10 coal, a U. S. limestone (Ref. BCR-1359) was used. Supplied by NAPCA, this had previously been used in related studies at the Argonne National Laboratories.

Table 3 is a summary of experimental conditions used for the tests described.

During all runs, samples of solids were collected and an extensive programme of analysis carried out to obtain mass balances for all important components. It is not proposed to discuss these in the present context except to note that in the earlier work there was frequently a discrepancy in the sulphur balances amounting to some 20

Table 1. DETAILS OF COALS

Coal % Compared (as received)	U.K.			U.S.A.		
	Goldthorpe	Welbeck	Park Mill	Arkwright	Farmington	(Argonne) Peabody No. 10
Moisture, %	5.0	4.2	2.1	2.1	1.8	9.1
Ash, %	17.0	18.2	16.5	13.9	9.8	11.4
V.M., %	29.7	29.7	31.9	34.4	28.3	38.2
F.C., %	47.7	47.9	49.5	49.6	50.2	41.3
Sulphur, %	2.05	1.25	2.4	2.25	2.25	4.1
Size Analysis						
+ 10	0.5	0.0	0.1	0.0	—	0.4
- 10 + 30	40	26	60	44	48	52
- 30 + 60	24	40	19	26	25	22
- 60 +120	17	17	10	15	14	13
-120 +240	9	7	5	7	8	7
-240	10	10	6	8	5	6

Table 2. PARTICLE SIZE OF BRITISH LIMESTONE

Sieve Size	As used with: Goldthorpe and Farmington	Arkwright	Welbeck, Park Mill, and Peabody No. 10
+ 10 B.S.	0	0.7	0.5
- 10 + 30	27	65	52.0
- 30 + 60	20	19.5	[illegible]
- 60 +120	18	9.3	10.0
-120 +240	19	4.6	7.5
-240	16	0.5	11.0

percent of the input. This was particularly noticeable at the higher levels of limestone addition. In the later tests (Welbeck, Park Mill, and Peabody No. 10), the better sulphur balances obtained were ascribed both to the greater attention that was paid to measuring the quantity of material in the bed at the beginning of the test period, and an improvement in sampling procedures for solids products.

Table 3. EXPERIMENTAL CONDITIONS INVESTIGATED

	Goldthorpe	Welbeck	Park Mill	Arkwright	Farmington	Peabody No. 10
Bed temp., °C	800	800	800	800	800	700 and 800
Bed height, ft	2	2	2	2	2	2 and 3
Fl. velocity, ft/sec	2	2 and 3	3	2	2	3
Recycle	Yes	Yes and No	No	Yes	Yes	No
Ca/S ratio	0-2.2	0-2.8	0-2.8	0-1.8	0-1.8	0-3.0

RESULTS

This paper considers only flue gas sulphur dioxide and the extent to which it is reduced by the addition of limestone under various test conditions.

Figure 2 plots the data. The zero condition is represented by the combustion of the coal without limestone addition; subsequent runs with limestone are plotted in terms of the percentage by which the SO_2 in the flue gas is reduced against the quantity of lime injected. This latter quantity, expressed as the Calcium/Sulphur (Ca/S) ratio, does not include the calcium natural to the coal. Thus, the figure reflects directly the effect of the limestone added and all data refers to a common origin, the "no limestone" condition.

The results with Welbeck coal, which has been used over the widest variety of experimental conditions, cannot be clearly represented in Figure 2. These are therefore shown in more detail in Figure 3, together with the Goldthorpe results already shown in Figure 2.

DISCUSSION

The results are discussed in terms of the following variables:

1. Quantity of limestone.
2. Quality of limestone.
3. Effect of fluidising velocity.
4. Effect of recycle.
5. Coal caking properties.
6. Coal sulphur content.

Quantity of Limestone

Considering all the data with the British limestone at a fluidising velocity of 3 ft/sec without recycle of fines, these form a group with the following average values:

Ca/S Ratio	SO_2 Retained %	
	Average	Range
1	48	40 - 54
2	75	68 - 83
3	94	90 - 10

At a Ca/S ratio of 2, the likely practicable maximum, the figure of 75 percent of SO_2 retained by the limestone means that 38 percent of the calcium was utilised. The results for the U. S. limestone with Peabody No. 10 coal shows 63 percent of SO_2 retained at a Ca/S ratio of 2 which is a calcium utilisation of 32 percent. This is poorer than the results with British limestone and is confirmed by the results at a Ca/S ratio of 3 which show a progressively poorer performance by the U. S. limestone. These differing results at high Ca/S ratios could imply small difference in the reactivity of the limestone but may be due to other causes; e.g., particle size.

Quality of Limestone

All of the limestones contain more than 97 percent calcium carbonate which removes one possible source of variation. The differences in particle size analysis for the various batches of U. K. limestone could also be a source of variation in performance. However, because the earlier tests on Goldthorpe coal using a finer limestone also used recycle, a direct comparison between the different U. K. limestone batches is not possible. The poorer performance of the U. S. limestone, detectable principally at high Ca/S ratios, may be due to its finer particle size compared with the equivalent batch of U. K. limestone. The tests concerned were all at 3 ft/sec without recycle and it is likely that under these conditions much fine limestone is lost from the system without being utilised. A further test using the U. S. limestone with recycle could resolve this point.

Effect of Fluidising Velocity

The tests with Welbeck coal provide the only direct evidence on this variable; from Figure 3 it is apparent that there is a small loss in efficiency caused by raising the fluidising velocity from 2 to 3 ft/sec. This amounts to about 10 percent of the input sulphur content for Ca/S ratios between 1 and 2. If this factor is applied to the Goldthorpe data over the same range of limestone/sulphur stoichiometry, then it could be predicted that Goldthorpe would still respond marginally better to limestone addition than did the other coals. However, it must be remembered that the limestone used for Goldthorpe was finer than for Welbeck.

Effect of Recycle

Again, the only relevant data is from Welbeck coal at 2 ft/sec; it is clear from Figure 3 that no effect was detected. This seems slightly improbable at first sight but, as the limestone was approximately 70 percent + 60 B.S. (250 micron), it could be argued that the small proportion of fine limestone lost by elutriation would not significantly help the sulphur retention figure even if it were recycled. The effect of recycle at higher fluidising velocities could be expected to be more marked.

Coal Caking Properties

The Goldthorpe coal is weakly caking and originally its superior performance to Farmington and Arkwright was thought to be at least partly on account of the highly caking properties of the two latter fuels. Welbeck (also a weakly caking coal) gave results closely similar to Farmington and Arkwright which appears to contradict this assumption. However, if a 10 percent bias is applied to the Arkwright and Farmington data, to equate them with the 3 ft/sec results, they then become closely comparable with Park Mill—a caking coal—and appreciably lower than the Welbeck results.

Thus, using a common limestone with all data "corrected" to 3 ft/sec, the caking coals fall into a compact group inferior in performance to the other coals tested. Developing a hypothesis to explain this behavior presents serious problems.

Arguments based on incomplete combustion leading to incomplete release of sulphur as SO_2 should lead to better than expected efficiencies in terms of flue gas SO_2 analysis. Agglomeration in the bed, leading to envelopement of limestone particles and hence their incomplete sulphation, is possible; however, no agglomerates have been observed in any of the experiments. Simple delay in the release of sulphur oxides from the bed is possible; however, since both Arkwright and Farmington were tested both with recycle of fines and with a fine limestone, there is plenty of opportunity for the recycled limestone to contact the sulphur oxides. However, it could be argued that by contrast to the situation within the fluid bed, the gas/solid contacts in the gas stream are insufficient to make up for lower sulphur retention in the bed. Further experiments using caking coals with and without recycle are needed to explore this suggestion more fully.

Coal Sulphur Content

There is no directly observable effect due to the total sulphur content of the fuel. Welbeck has the lowest sulphur content of the coals tested (1.25 percent a.r.) and the results for this coal correspond quite closely to those for the Peabody No. 10 coal which has the highest sulphur content (4.1 percent a.r.). Information currently available on the forms of sulphur in the coals does not lead to any meaningful correlation, but this is to be examined in more detail in other experiments.

CONCLUSIONS

At a fluidising velocity of 3 ft/second without recycle of fines, sulphur retentions of better than 90 percent can be achieved but require approximately three times the stoichiometric quantity of limestone.

An increase in fluidising velocity from 2 to 3 ft/second decreases sulphur retention by approximately 10 percent of the sulphur input. At these modest fluidising velocities the effect of recycle is small but may be increased significantly: at higher velocities, or with finer limestone.

A reduction in sulphur retention is apparently related to coal properties in that the more strongly caking coals are less responsive to limestone addition. An adequate explanation for this has yet to be developed and further tests are needed.

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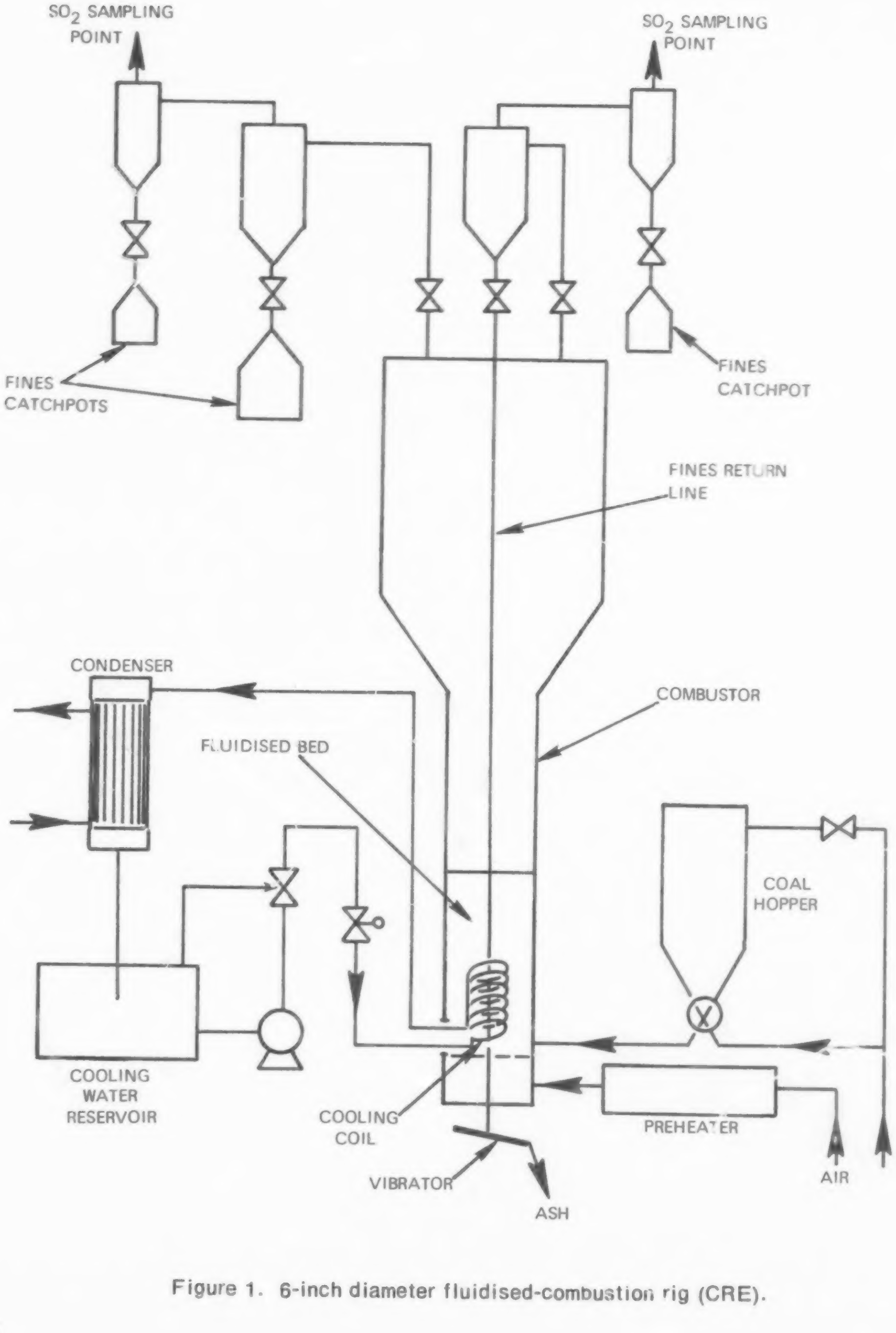

SO$_2$ SAMPLING POINT
SO$_2$ SAMPLING POINT
FINES CATCHPOTS
FINES CATCHPOT
FINES RETURN LINE
CONDENSER
COMBUSTOR
FLUIDISED BED
COAL HOPPER
COOLING WATER RESERVOIR
COOLING COIL
PREHEATER
VIBRATOR
ASH
AIR

Figure 1. 6-inch diameter fluidised-combustion rig (CRE).

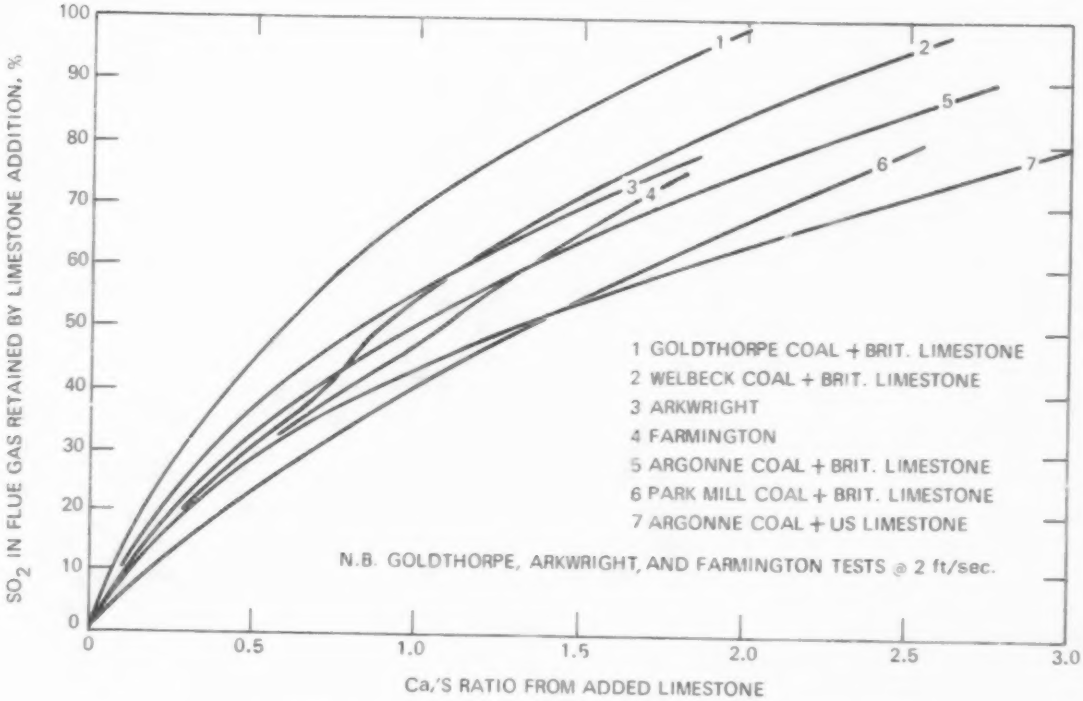

Figure 2. Effect of limestone addition on retention of SO$_2$.

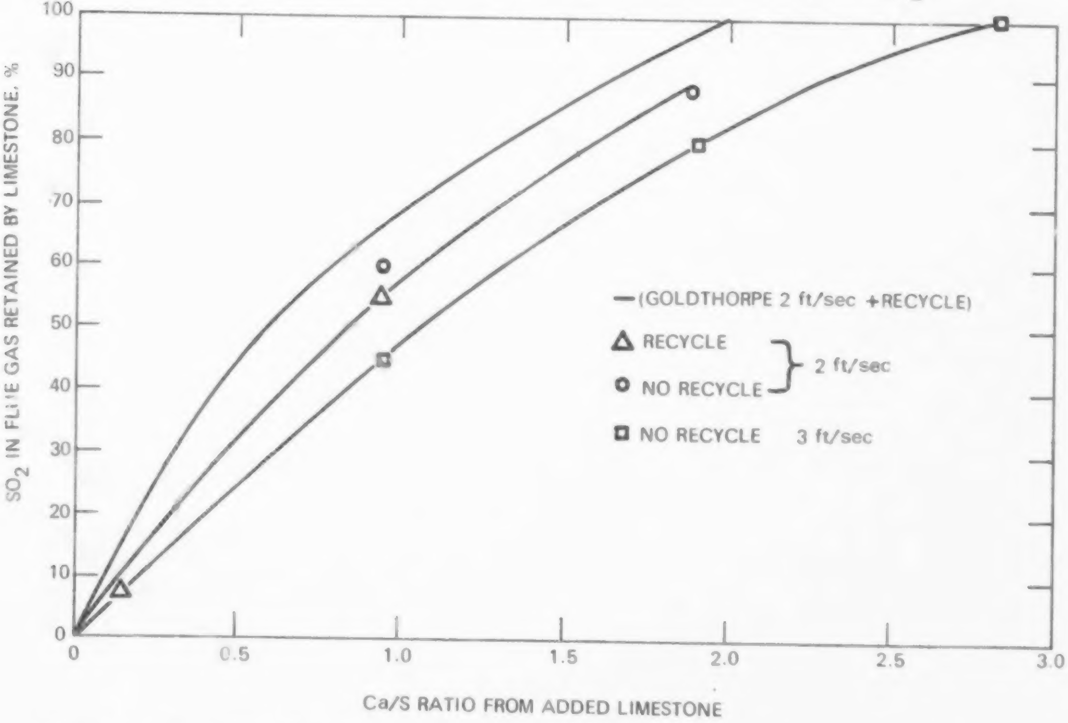

Figure 3. Retention of sulphur by limestone addition to the feed (Welbeck coal).

2. CHARACTERIZATION OF EMISSIONS FROM FLUIDIZED-BED COMBUSTION OF COAL AND CONTROL OF SULFUR EMISSION WITH LIMESTONE

R. D. GLENN AND E. B. ROBISON

Pope, Evans and Robbins

INTRODUCTION

This paper presents a brief review of emission studies in the area of fluidized-bed combustion, conducted by Pope, Evans and Robbins for the National Air Pollution Control Administration. The studies, in general, involved a characterization of emissions from fluidized-bed combustion of coal, sulfur emission control by limestone injection into the bed, and sulfur emission control by combustion of coal in a limestone bed. This paper also presents an estimate of the potential for sulfur emission control by continuous regeneration of a limestone bed system based on some preliminary experimental data.

CHARACTERIZATION OF EMISSIONS

The initial investigation to characterize the emission from the coal-fired fluidized bed indicated levels of sulfur dioxide, sulfur trioxide, nitric oxide, hydrocarbons, and particulates, and the extent to which these could be modified by purely operational changes; e.g., changes in excess air rate, coal feed rate, and bed temperature. A summary of the results of this investigation follows.

Sulfur Oxides

About 95 percent of the sulfur in the coal appeared as sulfur dioxide in the flue gas; the balance was retained by the ash. A very small percentage, 1 percent or less, appeared as sulfur trioxide in the flue gas. In the absence of a sorbent, the sulfur emission was substantially unaffected by changes in the operating variables.

Nitrogen Oxides

Emission of nitric oxide (NO) was found to be the dominant form in the NO_x group. The emission level, although low in comparison to the level of other coal-fired units, was still an order of magnitude greater than the equilibrium value predicted from the bed temperature. Furthermore, the NO level did not change when bed temperature was changed. This result led to the conclusion that NO emission was controlled by local temperatures higher than the measured bed temperature.

The NO emission responded only to increase in the oxygen content in the flue gas (increase in excess air). In one test an NO concentration of 280 ppm, observed at 1 percent oxygen in the flue gas, increased to 340 ppm at 4 percent oxygen. This variation is shown in Figure 1. The average concentration of a test series conducted at 3 percent oxygen in the full-scale unit was 275 ppm.

Hydrocarbons

The full-scale boiler could be operated with as little as 5 percent excess air without visible smoke in the flue gas; however, hydrocarbon concentrations were found to be as high as 1500 ppm (methane) at this excess air level. This high concentration was reduced sharply as the excess air rate was increased. Hydrocarbon burnup was essentially complete at 24 percent excess air (4 percent O_2 in the flue gas). The variation in shown in Figure 2.

Particulates

In the test system, particulate emissions were a function of the cyclone collector efficiency. Analysis indicated that 90 percent of the fly ash that passed the cyclone was smaller than 20 microns in particle size.

EFFECT OF OPERATIONAL CHANGES

Effect of Limestone Particle Size

When limestone (BCR 1359) was added to the bed in a comparatively coarse particle size (-7 +14 mesh), the sulfur absorption was rather poor. Under the most favorable conditions, the limestone utilized in the sulfur capture was limited to about 20 percent. Emission of SO_2 was reduced by about 20 percent at a stoichiometric ratio of 1.0 and about 40 percent at a stoichiometric ratio of 2.0. A dolomitic limestone (BCR 1337) of the same size was more effective when the stoichiometric feed ratio was based on the calcium fraction alone; however, on a total weight basis, it was equally poor.

At this point in time, it became evident to many researchers that utilization of sorbent was limited by the formation of a sulfate shell on the surface of the sorbent particle. Improvement required an increase in the surface-to-mass ratio of the particle; i.e., a reduction in particle size.

When limestone was ground to a fine powder (-325 mesh) and injected into the bed, the sulfur capture was markedly improved. The reduction in SO_2 emission with increase in stoichiometric rates is shown in Figure 3. The trend shows SO_2 reductions of 40, 66, and 84 percent at respective Ca/S ratios of 1, 2, and 3. The corresponding limestone utilizations are 40, 33, and 28 percent. These results were obtained with a high sulfur (4.5 percent) coal burning in a 10- to 12-inch deep fluidized bed operating in the temperature range of 1500 - 1600°F, and with a superficial velocity of 12-14 ft/sec. Comparable results were observed with a medium (2.6 percent) sulfur coal.

The lower line on Figure 3 shows the greater reactivity obtained with a dolomitic limestone; however, the stoichiometric ratios are calculated for calcium only. If the weight of magnesium is included and results are expressed in practical terms, such as pounds of stone fed per pound of sulfur removed, dolomite has no advantage over limestone.

In a short series of tests with calcined limestone, relatively poor results were obtained, as indicated by the upper line of Figure 3.

A detailed study with the particular limestone used (No. 1359) showed that fine grinding would be necessary for favorable sulfur capture. This limestone is one of the most durable, however, and such fine grinding may not be necessary with less durable limestones which may decrepitate from thermal shock during the initial calcination stage.

Figure 4 shows the effects of limestone particle size, bed depth, and temperature on SO_2 reduction. All of the data in this series was obtained while burning a 3-percent sulfur coal in a fluidized bed of sintered ash. Limestone was injected at a stoichiometric ratio of 2.6.

At particle sizes below about 400 microns, the beneficial effects of reducing the particle size of the limestone injected are clearly evident. At particle sizes above 400 microns, some other factor—probably increased residence time in bed—more than offsets the decrease in surface area, and a slight improvement is obtained with increase in particle size. From a practical viewpoint, however, only high reductions are important.

If SO_x emissions from a fluidized-bed boiler are to be controlled by the injection of limestone, it must be a very finely divided limestone.

Effects of Bed Depth and Temperature

Figure 4 also shows the effect of bed depth. As expected, an 18-inch bed shows a higher absorption than does a 10-inch bed.

Also shown is the effect of bed temperature; the beneficial effects of operating at a temperature of 1550°F are clearly indicated. The poorer results obtained at 1675°F and at 1800°F bring to mind some of the poor results that have been obtained by injecting powdered limestone into a conventional pulverized-coal-fired furnace which must operate at even higher temperatures. One of the outstanding advantages of fluidized-bed combustion is its ability to operate economically at temperatures of 1500 to 1600°F. This makes the process particularly well adapted for sulfur emission control through the use of limestone.

LIMESTONE INJECTION

The injection of limestone for sulfur control had no observable effect on hydrocarbon emission. In several instances the addition of limestone reduced NO emission but the effect was not reproducible. Sulfur trioxide emission was reduced to zero whenever limestone was injected.

Results of the limestone injection tests indicate that a 4.5-percent sulfur coal can be made the equivalent of a 1-percent sulfur coal with the injection of 1359 limestone at a rate of 27 lb/1000 lb of coal. A 2.6-percent sulfur coal can be converted to a 1-percent sulfur equivalent with the addition of 1359 limestone at a rate of 10 lb/100 lb of coal. The corresponding Ca/S ratios are 1.9 and 1.2.

LIMESTONE BED

As an alternative to injecting pulverized limestone, limestone was used as a bed material. The effect on SO_2 emission was remarkable.

Tests conducted with a limestone bed of -10 +20 mesh particle size and a 3-percent sulfur coal indicated that SO_2 emission could be almost completely eliminated for a period of 2 to 3 hours. After this period, SO_2 began to appear in the flue gas above the bed with concentrations increasing with time. After about 4 hours, the SO_2 emission represented about 30 percent of the input and the concentration of sulfur in the bed rose to 7.4 percent. This concentration corresponds to a limestone utilization of about 16 percent. A once-through process in a limestone bed was clearly uneconomical, and attention was given to the regeneration of the limestone for reuse. Emissions monitored during both adsorption and regeneration are shown in Figure 5.

In this test the sulfur content of the coal was again 3 percent, corresponding to a flue gas concentration of 0.24 percent SO_2. As before, for the first 2 hours, substantially all of the SO_2 was removed. The concentration in the flue gas then rose gradually and, after a little over 3 hours, the SO_2 emission rose to about a third of what it would have been if coal ash rather than limestone had been used as the bed material.

The operating conditions were then changed. The coal rate was increased so that the temperature, which had been 1550°F, increased to 1950°F. The oxygen concentration in the flue gas, which had been 3 percent, was dropped to 1 percent. A rapid desorption of SO_2 took place, and 90 percent of that which had been absorbed during more than 3 hours of operation, was desorbed in a few minutes. The SO_2 concentration of the flue gas reached a peak of 8.1 percent–a value which is more than 30 times higher than the 0.24 percent that would have been experienced if the coal had been burned without the use of limestone in the bed.

REGENERATIVE PROCESS

The results observed in this test suggested a regenerative process for using a limestone bed to control SO_2 emissions from a fluidized-bed boiler. Absorption might be carried out in the fluidized bed of the main part of the boiler. Desorption, and the production of a flue gas that is very high in SO_2 concentration, may be carried out in another section. The product shell limitation–the mechanism by which absorption of SO_2 on the surface of a limestone particle reduces its effectiveness for the absorption of additional SO_2–can be circumvented by repetitive use of the particle surface. The high concentration of SO_2 in the small volume of flue gas from the desorption section can be removed and/or recovered by any of the variety of processes that have previously been proposed for the treating of conventional boiler flue gas.

These processes, presently high in cost, will become economical as a result of the high SO_2 concentration and low volume of flue gas to be treated.

A patent application has been filed on this "SO_2 Acceptor Process."

The performance of a continuous absorption and regeneration system has been estimated from the SO_2 removal efficiency of the regenerated limestone bed. Experimental observations of SO_2 emission after regeneration of the bed showed that the emission rate increased linearly with time. The total sulfur in the bed also increased simultaneously in a linear fashion. The rate of sulfur emission could, therefore, be related to the total sulfur in the bed for a system operating with a 3.0-percent sulfur coal.

A sulfur balance around an absorption cell employing recycle of regenerated limestone follows the relation:

$$L \frac{dx_s}{dt} = G_0 + (x_0 - x_s)\, L\, f_L - G_e \qquad (1)$$

where L is the bed mass, x_s the sulfur fraction in the bed, t time, G_0 the sulfur input rate, x_0 the sulfur fraction in the regenerated limestone, f_L the fraction of the bed recycled per unit time, and G_e the flue gas sulfur emission rate.

Relating G_e to the sulfur in the bed, equation (1) becomes:

$$L \frac{dx_s}{dt} = G_0 + (x_0 - x_s)\, L\, f_L - K x_s L \qquad (2)$$

where K is the proportionality constant between the gas sulfur emission rate and the total sulfur in the bed.

Integrating equation (2), the sulfur fraction of the bed varies as follows:

$$x_s = \frac{G_0 + x_0 L f_L - [G_0 + x_0 L f_L - x_0 (KL - L f_L)\,]\, e^{-(K + f_L)t}}{KL + L f_L} \qquad (3)$$

The transient term indicates that x_s approaches equilibrium rapidly as the recycle rate is increased.

At equilibrium, the transient term disappears and

$$x_s = \frac{G_0 + x_0 L f_L}{L(K + f_L)} = \frac{G_e}{KL} \qquad (4)$$

If the sulfur fraction of the regenerated limestone (x_0) is small, the term $x_0 L f_L$ can be neglected. The sulfur emission rate G_e is then related to the sulfur input rate, the gas constant, and bed recycle rate by the relation:

$$G_e = \frac{K(G_0)}{K + f_L} \quad \text{or} \quad \frac{G_e}{G_0} = \frac{K}{K + f_L} \qquad (5)$$

The value of K determined by experiment was found to be 0.32 hr^{-1} (lb sulfur emission per hr per lb sulfur in the bed).

The measured values of $\frac{dG_e}{dt}$, $\frac{dx_s}{dt}$, and L were 0.37 lb/hr^2, 0.0215 hr^{-1}, and 55 lb, respectively.

Figure 6 is a plot of the calculated SO_2 removal versus the number of bed changes per hour. It is indicated that, with only one bed change per hour, a 76-percent reduction of SO_2 emissions may be obtained. With two bed changes per hour, an 87-percent reduction is predicted, and with three bed changes per hour, a 91-percent reduction is predicted.

These results indicate a very favorable potential for sulfur emission control by limestone regeneration in a continuous operation. Further investigation is planned.

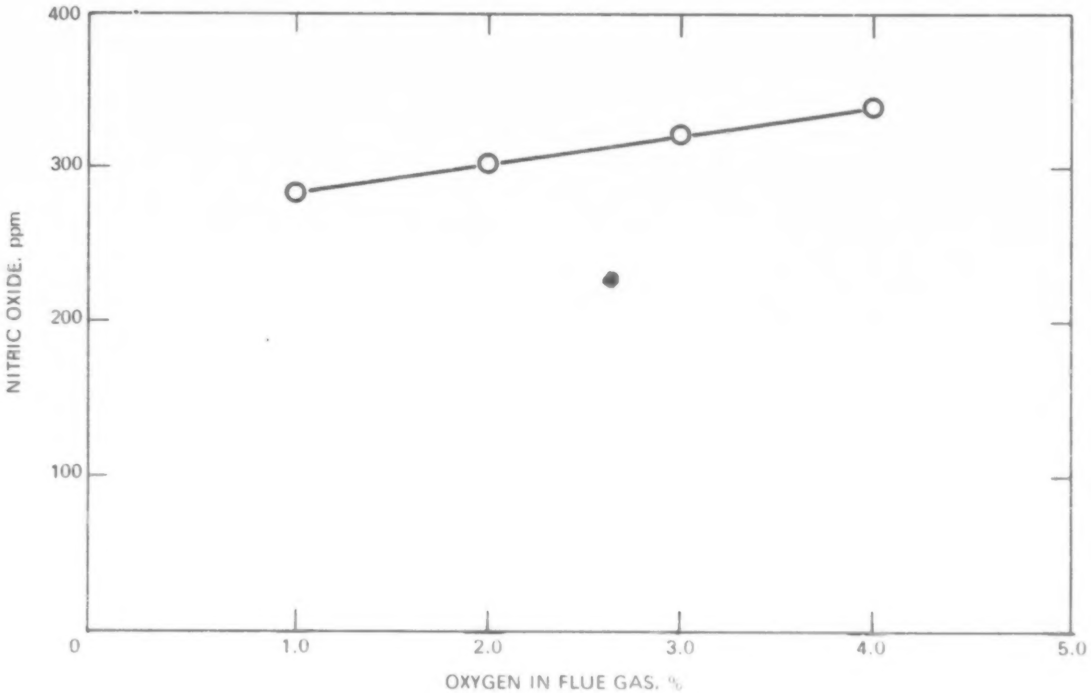

Figure 1. Effect of oxygen concentration on NO emission.

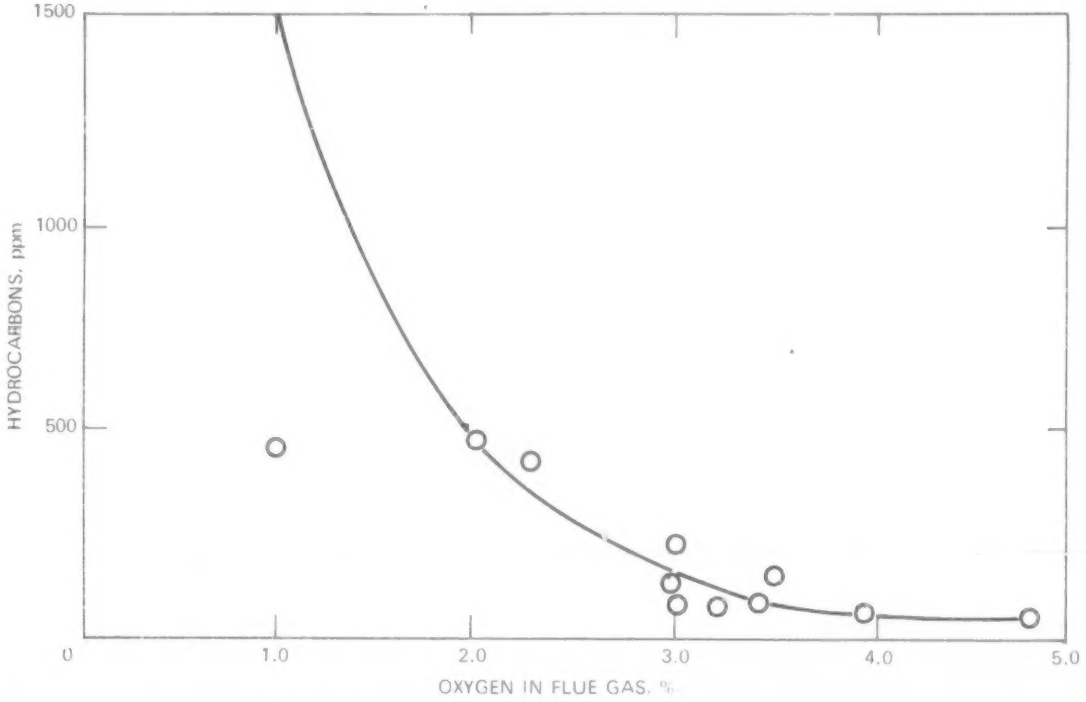

Figure 2. Effect of oxygen concentration on hydrocarbon emission.

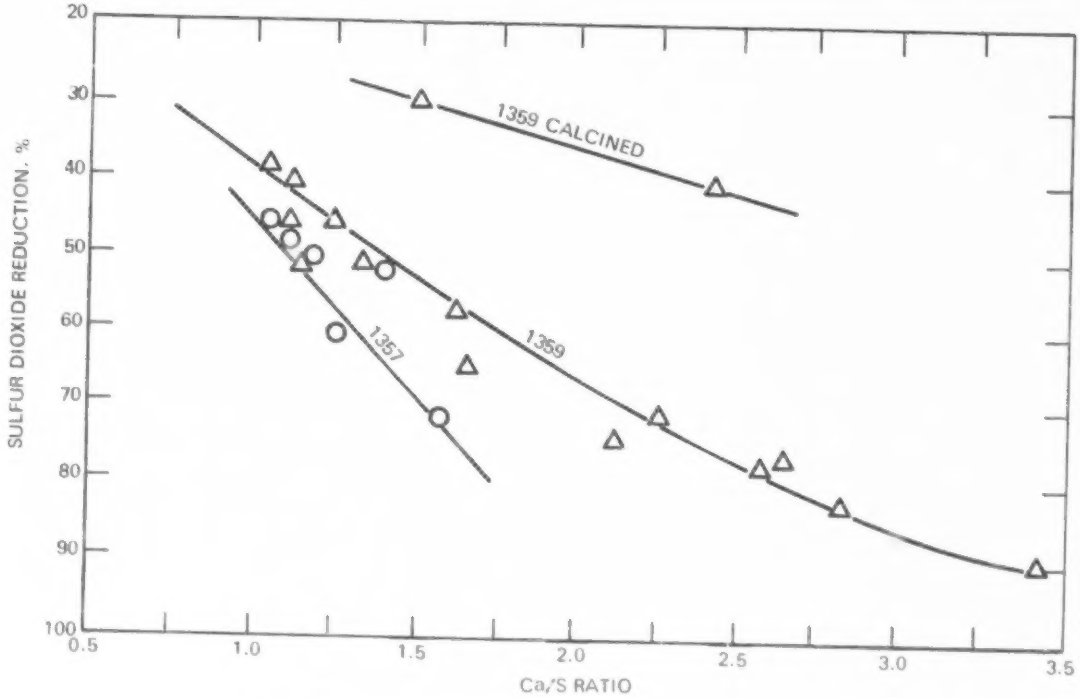

Figure 3. SO_2 reduction - - effect of limestone-to-sulfur ratio.

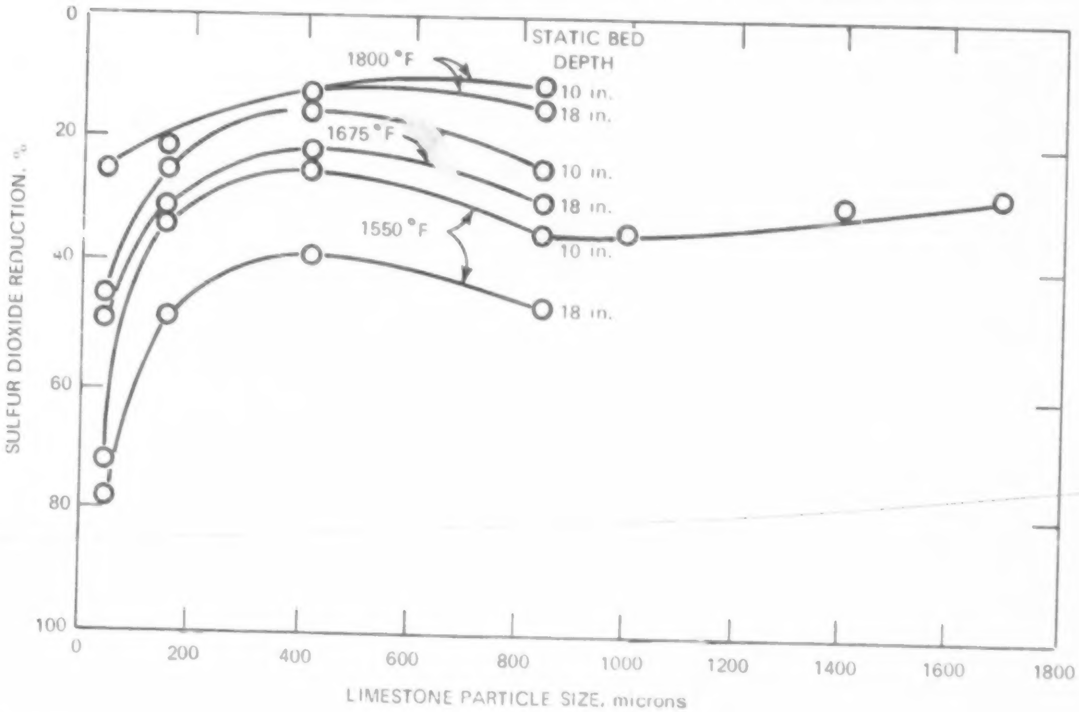

Figure 4. SO_2 reduction - - effect of limestone particle size, bed depth, and temperature.

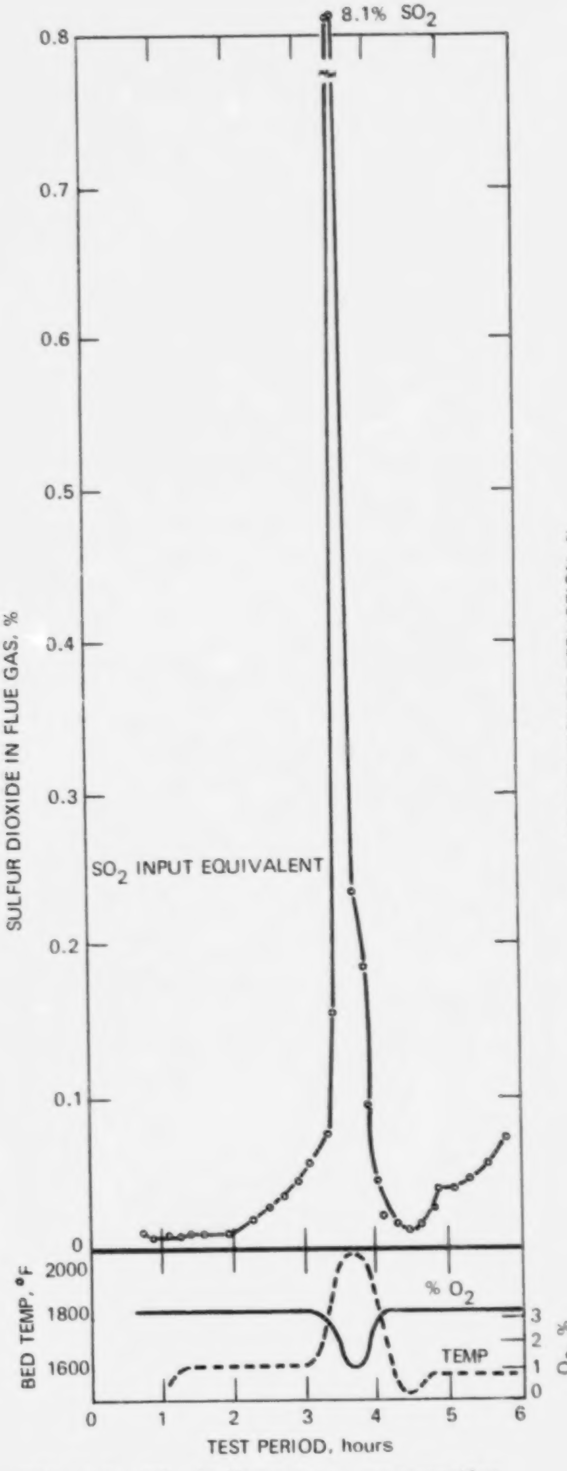

Figure 5. SO_2 in flue gas - - regenerative process using a limestone bed.

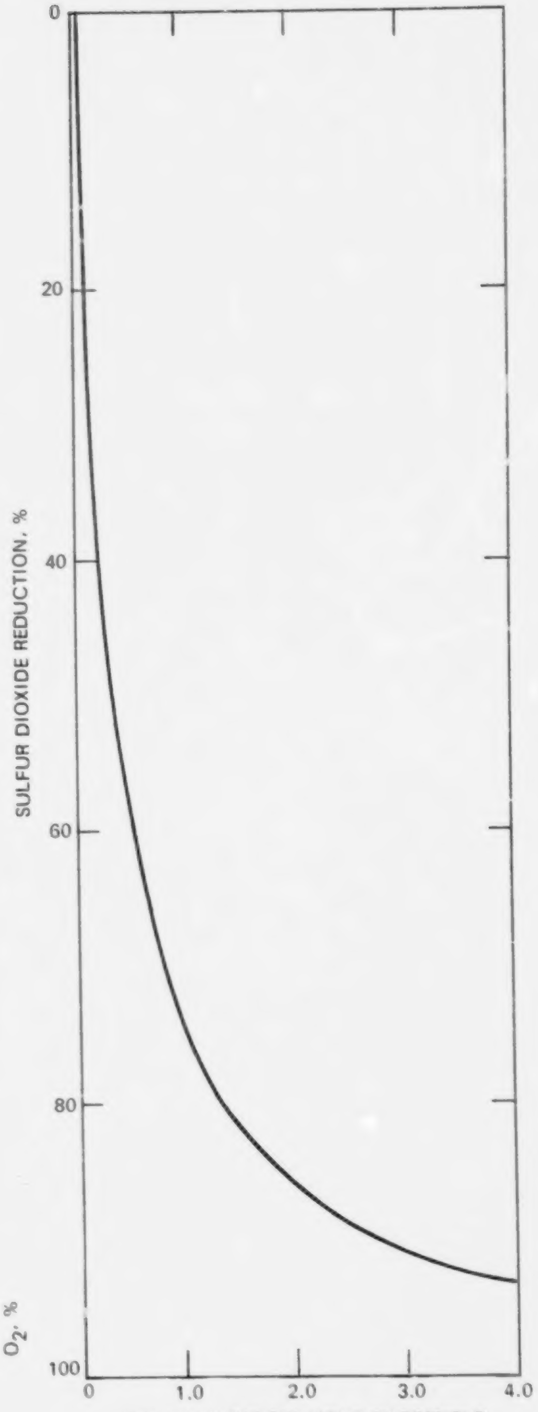

Figure 6. Calculated SO_2 removal by regenerative process.

3. POLLUTION CONTROL CAPABILITIES OF FLUIDIZED-BED COMBUSTION

L. J. ANASTASIA, E. L. CARLS,
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Argonne National Laboratory

ABSTRACT

The combustion of a high-sulfur coal has been studied in a 6-inch dia fluidized-bed reactor. Basic additives containing CaO (e.g., limestone and dolomite) have been used for SO_2 control during combustion. The parameter having the greatest effect on SO_2 emission has been found to be the Ca/S ratio in the additive and coal feed streams. At Ca/S mole ratios of 2.0 to 2.5, about 80 to 90 percent of SO_2 emission can be prevented. When coarse additive particles are used as both the fluidized bed and additive, the optimum temperature range for SO_2 removal appears to be 1480 to 1550°F.

INTRODUCTION

Fluidized-bed combustion is being studied at Argonne National Laboratory as a method of reducing the quantity of atmospheric pollutants (oxides of sulfur and nitrogen) released during the combustion of fossil fuels. This work is being carried out under contract with the National Air Pollution Control Administration.

The concept of fluidized-bed combustion involves burning fuel in a fluidized bed of solids. In an industrial application, boiler tubes would be immersed in the bed. The rapid motion of the fluidized particles at the heat transfer surfaces, together with high heat-transfer rates between gas and particles, makes the fluidized bed a highly efficient heat-transfer medium. If the excellent heat transfer characteristics of fluidized-bed combustors allow combustor size to be decreased, lower capital and operating costs than for conventional units may be expected. Another advantage of the fluidized bed in this application is that it is a highly efficient contacting medium for carrying out gas-solids reactions; therefore, use of a bed material that reacts with gaseous pollutants generated during combustion offers good prospects for controlling air pollution. Also, combustion can be carried out at lower temperatures in a fluidized bed than by conventional combustion methods.

Disadvantages of the fluidized bed include a gas velocity limited by the extent of entrainment of unburned carbon from the fluidized bed; therefore, relatively large bed areas and a large number of fuel introduction points are required. The rapid motion of the fluidized particles results in some decrepitation of bed particles; the viscous drag of the gas is sufficient to entrain fine particles from the reactor, necessitating off-gas cleanup of both additive and fly ash particles.

To control sulfur oxide emissions, limestone or dolomite additive may be fed continuously to the fluidized-bed combustor, where calcination to lime (CaO) occurs simultaneously with combustion of the fuel and in situ reaction of sulfur oxides with lime to form calcium sulfate.

Two alternative modes of operation for a desulfurizing fluidized-bed combustor have been considered in which the major differences pertain to additive particle size and composition of the fluidized bed. In both modes of operation, crushed and washed coal (-6 and -14 mesh) was used; at the fluidizing-gas velocities employed (3 and 9 ft/sec), essentially all the coal ash generated by combustion of these coal particles was elutriated from the fluidized bed. In the first mode of operation, pulverized additive (<325 mesh) was used for SO_2 control, and the fluidized bed was refractory alumina having a relatively coarse particle size (30 mesh). Since most of the ash and additive was elutriated from the bed during a run, the fluidized bed at steady state was composed largely of alumina particles. In the second mode of operation, large additive particles used for SO_2 control were of sufficient size to remain in the fluidized bed. At steady state in this mode of operation, the fluidized bed was composed essentially of partially sulfated additive material.

EQUIPMENT, PROCEDURE, AND MATERIALS

Figure 1 is a schematic diagram of the fluidized-bed combustor system. The combustor is a 6-inch dia stainless steel vessel equipped with electric heaters and an air cooling system for temperature control. To start a run, preheated air is passed into the combustor through a bubble-cap air distributor mounted on the bottom flange. Electric heaters raise bed temperature to 1000°F, at which point coal is fed to the bed at controlled rates. Ignition of the coal increases the bed temperature to the desired operating temperature (i.e., ∿1600°F). When combustion conditions have stabilized, feeding of additive and/or recycle of elutriated fly-ash/limestone mixture is started. Variable-drive volumetric screw feeders mounted on scales allow solids feed rates to the system (coal, additive, and recycled solids) to be metered. The solids are fed pneumatically to the fluidized bed at a point just above the gas distributor.

Flue gas from the combustor is passed through two high-efficiency cyclone separators in series and a fibrous-glass final filter to remove entrained solids. Downstream from the cyclones, approximately 5 percent of the total flue gas is diverted to a gas analysis system. The water content of the flue gas sample is reduced to 3000 ppm (by condensation and refrigeration) to prevent the moisture from interfering with gas analysis. Continuous analyses of the dried gas for NO, SO_2, and oxygen are carried out by infrared analyzers and a paramagnetic oxygen analyzer. Gas chromatography provides intermittent analyses for CO_2. Periodically, the bed and overhead solids are sampled for chemical analysis and to obtain material balances.

A Central Illinois bituminous coal mined in Christian County has been used in all experiments reported here. The coal contains 4.5 wt percent S and 12.3 wt percent ash and has a heating value of 11,550 Btu/lb. The additive materials studied include:

1. Limestone No. 1359, Stephens City, Virginia (97.8 wt percent $CaCO_3$, 1.3 wt percent $MgCO_3$).
2. Limestone No. 1360, Monmouth, Illinois (78.0 wt percent $CaCO_3$, 22.0 wt percent $MgCO_3$).
3. Dolomite No. 1337, Gibsonburg, Ohio (53.4 wt percent $CaCO_3$, 46.5 wt percent $MgCO_3$).
4. Tymochtee dolomite, Huntsville, Ohio (49.3 wt percent $CaCO_3$, 36.6 wt percent $MgCO_3$).

In several experiments with limestone No. 1359, the fuel was natural gas supplied by a public utility through commercial gas lines.

Table 1 summarizes operating conditions and variables studied in combustion experiments. (Experiments pertaining to NO

Table 1. OPERATING CONDITIONS AND VARIABLES STUDIED IN FLUIDIZED-BED COMBUSTION EXPERIMENTS

Fuel	Number of Experiments	Starting Bed Material	Additive		Combustion Temperature, °F	Superficial Gas Velocity, ft/sec	Variables Studied
			Type	Average Particle Size, μm			
Coal	9	Alumina	Limestone No. 1359	25	1600	3	a) Gas velocity
				600	1600	3	b) Recycle of elutriated solids
				1400	1600	9	c) Additive particle size
Coal	4	Alumina	Limestone No. 1359	25	1550-1650	3	a) Combustion temperature
				103			b) Additive particle size
							c) Ca/S mole ratio
Gas	3	Alumina	Limestone No. 1359	25	1600	3	a) SO_2 removal with natural gas combustion
							b) Additive regeneration
Coal	4	Alumina	Tymochtee dolomite	<44	1600	3	a) Additive type
							b) Additive particle size
							c) Ca/S mole ratio
Coal	5	Partially sulfated limestone	Limestone No. 1359	615	1600		a) Limestone fluidized bed
			Limestone No. 1360	530	1600		b) Combustion temperature
			Dolomite No. 1337	540	1480-1800		
Coal	1	Partially sulfated limestone	Limestone No. 1359	490	1400-1600	3	a) Combustion temperature

removal, discussed in another paper, are omitted from this table.) The variables studied include gas velocity, recycle of elutriated solids, combustion temperature, additive type, particle size, fluidized-bed material, and the Ca/S mole ratio in the feed streams. As discussed below, the Ca/S mole ratio in the additive and coal feed streams appears to have the greatest effect on SO_2 control.

SO_2 EMISSION CONTROL WITH FINE-PARTICLE ADDITIVE

The most thoroughly studied additive for SO_2 control has been limestone No. 1359 because:

1. It has a minimum amount of waste burden (e.g., its $MgCO_3$ content is 1.3 wt percent).

2. In preliminary tests[1] this stone showed a high chemical reactivity with SO_2.

Pulverized limestone No. 1359, with an average particle size of 25 μm, was studied at addition rates equivalent to Ca/S mole ratios in the feed up to ∿3; Figure 2 summarizes the results. The SO_2 content of the flue gas was reduced by about 80 percent at a Ca/S mole ratio of 2.5.

Figure 2 includes SO_2 removals for runs in which recycled solids (a mixture of partially sulfated limestone and fly ash) were introduced into the combustor along with the additive and coal feeds. For runs in which recycled material was added, the contribution of unsulfated calcium in the recycle material was not included in the calculated Ca/S ratio. The absence of any improvement in SO_2 removal, when using fresh limestone plus recycled fly-ash/additive mixture, is puzzling since the recycled solids contain significant quantities of unreacted CaO. Also, as discussed below, separate laboratory tests on the reactivity and capacity of fresh limestone and the recycle material detected no significant difference in reactivity for the two materials.

The effect of superficial gas velocity (3 and 9 ft/sec) for limestone No. 1359, with an average particle size of 25 μm, is shown in Figure 3. At the higher gas velocity, there was less SO_2 reaction with fine additive particles. The difference is not large but does appear to be significant; this result is not unexpected since at the higher gas velocity, elutriation rates and, therefore, bypassing of solid particles are increased. At a superficial gas velocity of 3 ft/sec, the residence time for additive particles, calculated from the weight of the fluidized bed and the additive feed rates, is about 1 hour; however, experimental evidence indicates that, even at the low gas velocity, actual residence time for the bulk of the additive particles is much less.

The effect of combustion temperature in the range 1550 to 1650°F for 25-μm limestone No. 1359 particles (Figure 4) appears to be small; the data indicates that SO_2 removal at 1600°F is slightly better than at either 1550 or 1650°F. Figure 4 also shows that a 90-percent reduction in SO_2 emission occurs at a Ca/S mole ratio of ∿4.

The overall utilization (conversion of CaO to $CaSO_4$) of 25-μm limestone No. 1359 ranged from 25 to 40 percent (Figure 5). This utilization was calculated from the relationship between SO_2 removal and the Ca/S mole ratio in the feed streams. As might be expected from the relationship between Ca/S ratio and maximum CaO utilization (solid curve, Figure 5), higher utilization of CaO was obtained at the lower Ca/S ratios.

Chemical analyses of various effluent solids were performed. Significant differences in CaO utilization and in the extent of calcination for the different solids streams were noted. The results (Figure 6) show that material collected in the primary cyclone, which had the lowest extent of calcination, also had the lowest CaO utilization. Material retained in the fluidized bed, which had the highest extent of calcination, also had the highest extent of CaO utilization. Secondary cyclone solids were intermediate with respect to both extent of calcination and CaO utilization. Since most of the entrained solids in the flue gas might be expected to have similar residence times in the reactor, the difference in CaO utilizations for materials from the primary and secondary cyclones can be attributed to particle size. Larger particles were collected in the primary cyclone; finer particles, collected in the secondary cyclone, had a higher specific surface area and reacted more quickly and more thoroughly than the larger particles collected in the primary cyclone. For additive particles retained in the bed, residence times are long and a high degree of utilization and calcination can be achieved. The Ca/S mole ratios for cyclone samples and fluidized-bed samples taken during a single experiment with 25-μm limestone No. 1359 are shown in Figure 7. In the fluidized-bed samples, the Ca/S mole ratios were relatively constant at 1.5, which represents a CaO utilization of ∿70 percent. CaO utilization of elutriated material collected in the secondary cyclone ranged from 30 to 50 percent; CaO utilization for material in the primary cyclone was 20 percent or less. The data in Figures 6 and 7 suggests that calcination of $CaCO_3$ and utilization of the resulting CaO are dependent upon the same factors, which include particle size and residence time in the combustor.

In an attempt to increase the utilization of CaO, elutriated solids were recycled in the fluidized-bed combustor in several experiments. Material collected in the primary cyclone, having a CaO utilization of 20 percent, was recycled. As indicated in Figures 2 and 5, SO_2 removal was not discernibly improved by adding recycled material. To explain these results, laboratory tests were carried out to compare the rates of sulfation of fresh limestone and recycled solids. Figures 8, 9, and 10 show the results of these tests.

The reaction rates for fresh limestone and for recycled solids (mixture of fly ash and partially sulfated limestone) were essentially the same, both at the start of the experiment and after 2 hours of reaction (Figure 8). These tests were performed at an SO_2 partial pressure of 3.2 mm Hg, which is considerably higher than SO_2 pressures in the combustor (<0.8 mm Hg). Additional tests were performed (Figure 9) at an SO_2 partial pressure of 0.4 mm Hg; again, the reaction rates for the fresh limestone and the recycled solids were similar, both initially and after 2 hours of reaction. Thus, the apparently lower reactivity of the recycled fly-ash/limestone mixture in the fluidized-bed combustor cannot be attributed to a difference in reaction rate as a function of SO_2 partial pressure. The percentage of CaO utilized during the laboratory sulfation tests was determined for recycled solids and fresh limestone (Figure 10). As might be expected from the similarity of reaction rates, the total percentage of CaO utilized was the same for both materials. Thus, the ineffectiveness of recycle operation in reducing SO_2 levels cannot be accounted for by a reason intrinsic to the reaction of recycled material with SO_2.

Electron microprobe studies[2] were carried out in an attempt to explain the anomalous behavior of the recycled solids in the fluidized-bed combustor. Cross sections of typical particles, collected in the primary cyclone during combustion experiments with natural gas and with coal, were examined for homogeneity of sulfur and calcium content. Figures 11 and 12 show the results of the examinations. Some differences in particle composition noted are related to the fuel used. With coal as the fuel, sulfur concentration usually decreases at increasing distance from the surface of the particle; on the other hand, sulfur concentrations in particles collected during the combustion of natural gas appear to be more uniform throughout the particle. In gas combustion experiments, SO_2 was introduced into the coal feed port from an external source and SO_2 concentration in the fluidized bed may have been more uniform than during the combustion of coal. Three of the four particles from the gas experiment (Figure 12) had a calcium content quite close to that expected for $CaCO_3$; only one particle had a calcium and sulfur content expected for $CaSO_4$.

The laboratory tests have indicated that high utilization of the limestone is possible and that the reactivity of recycled solid is essentially the same as that of fresh limestones. A lack of further reaction of recycled solids in the combustor may be due to insufficient gas-solids contacting which, in turn, may result from short residence times. When both recycled solids and fresh limestone are fed to the combustor, the conditions in the combustor may favor greater initial reactivity of fresh limestone. Perhaps better utilization of the recycle material could be achieved with separate combustor beds for recycled additive and fresh additive feed.

Effect of Additive Material

The effectiveness of pulverized Tymochtee dolomite was compared with that of limestone No. 1359 for SO_2 control during coal combustion in a fluidized bed of alumina. Figure 13 shows that the difference between the additives is small, but that Tymochtee dolomite yields higher SO_2 reductions at Ca/S mole ratios smaller than 2. It is generally thought that the MgO content of calcined dolomitic additives helps keep the particle pore structure open so that more of the CaO can react with SO_2. However, the presence of MgO in dolomites is a serious disadvantage because MgO does not react with SO_2 and greatly increases the waste burden for dolomitic additives; e.g., the stoichiometric quantity to react with the sulfur in 1 lb of 4.5 wt percent S coal is 0.29 lb Tymochtee dolomite, compared with only 0.15 lb limestone No. 1359.

The waste burden for any of the additive materials can be considerably reduced in operational schemes which include an additive regeneration step. In such a scheme, sulfur values could be recovered from the spent additive, and regenerated additive can be utilized for SO_2 emission control so that the net waste burden of the system would only be due to fresh makeup additive.

SO_2 EMISSION CONTROL WITH COARSE-PARTICLE ADDITIVE

Experiments were performed with additive particles too coarse to elutriate from the fluidized bed; fluidized beds of partially sulfated additive of a composition close to that expected at steady-state operation were used. Table 2 shows the experimental results for limestones No. 1359 and 1360, and dolomite No. 1337. Data for coarse limestone is compared with results for pulverized limestone No. 1359 (25 μm) in Figure 14. The results for coarse additive particles tend to cluster in a region of small area, indicating a relative independence of the type of additive material tested and, for limestone No. 1359, a relative independence of particle size up to 615 μm. Although the coarse additive particles have lower reactivity toward SO_2 than do fine particles, their increased residence time and the resultant large mass of material in contact with the gas stream resulted in the same degree of SO_2 removal as with finer (more reactive) particles, which elutriate readily from the fluidized bed and hence have shorter contact times with the gas stream. The observed similarity of SO_2 removals for a variety of stones and particle sizes may be fortuitous or may indicate that the Ca/S mole ratio in the feed streams is highly significant.

The effect on degree of SO_2 removal of combustion temperature in the range 1400 to 1800°F was determined for coarse limestone No. 1359 and dolomite No. 1337 (Figure 15). The maximum SO_2 removal of ∼90 percent

Table 2. SO_2 REMOVALS[a] ACHIEVED WITH COARSE-PARTICLE ADDITIVE MATERIALS

Additive			
Material	Average Particle Size, μm	Ca/S Mole Ratio	SO_2 Removal, %
Limestone No. 1359	615	2.6	79
Limestone No. 1359	615	2.3	83
Limestone No. 1359	615	2.5	79
Limestone No. 1359	490	2.5	86
Limestone No. 1360	630	2.3	74
Dolomite No. 1337	540	2.2	81

[a] At combustion temperature of 1600°F; with gas velocity of ∿3 ft/sec.

occurred in the temperature range 1480 to 1550°F. In contrast, for pulverized limestone No. 1359 (Figure 4), SO_2 removal was little affected by variation of combustion temperature in the range 1550 to 1650°F. The different behaviors of coarse and fine limestone may be explained by recognizing that reaction of the pulverized limestone with SO_2 depends on the extent of calcination. Although finely powdered limestone readily elutriates from the bed, it has a high specific surface area and reacts at high rates so that the extent of calcination may be relatively independent of bed temperature in the range 1550 to 1650°F. Thus the optimum temperature for reaction with SO_2 from pulverized limestone is not necessarily the same as that for coarse limestone.

CONCLUSIONS

Control of SO_2 emission by reaction at 1400 to 1800°F with basic additives (limestone or dolomite) containing CaO has been studied in a 6-inch diameter fluidized-bed combustor. In the combustion of a high-sulfur Illinois coal, SO_2 emissions have been controlled effectively by feeding additive into the combustor. For each of the additives tested, Ca/S mole ratio in the feed had the greatest effect on SO_2 emission. Emission of about 80 to 90 percent of the sulfur has been prevented by feeding additive and coal at a Ca/S mole ratio of 2.0 to 2.5. For fluidized beds of limestone No. 1359 or dolomite No. 1337, the optimum combustion temperature for SO_2 removal appears to be 1480 to 1550°F. Variables that have minor effects on SO_2 removal are additive type, additive particle size, and superficial gas velocity.

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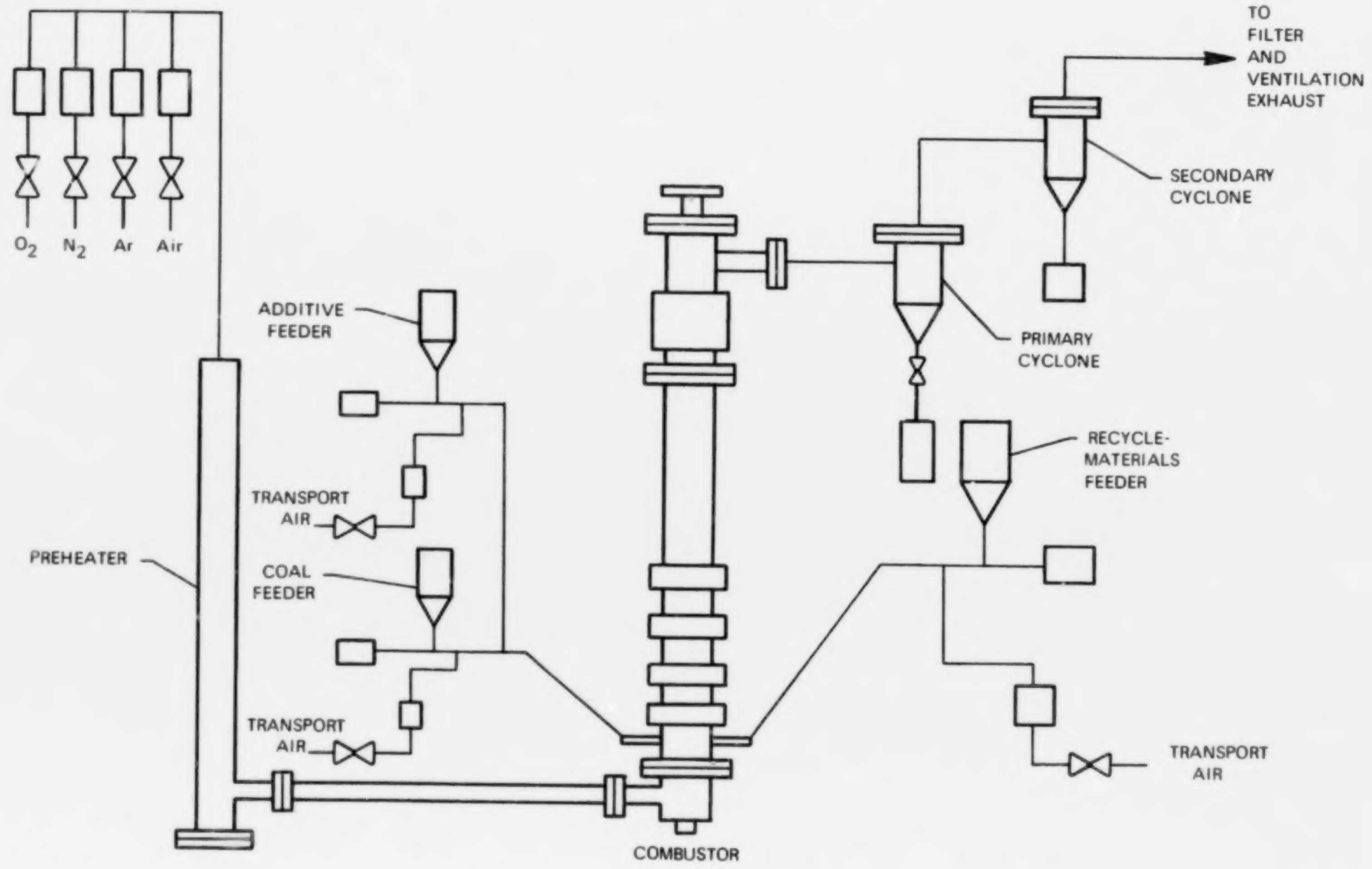

TO
FILTER
AND
VENTILATION
EXHAUST
SECONDARY
CYCLONE
O_2
N_2
Ar
Air
ADDITIVE
FEEDER
PRIMARY
CYCLONE
RECYCLE-
MATERIALS
FEEDER
TRANSPORT
AIR
PREHEATER
COAL
FEEDER
TRANSPORT
AIR
TRANSPORT
AIR
COMBUSTOR

Figure 1. Equipment for investigating SO_2 emission in combustion experiments.

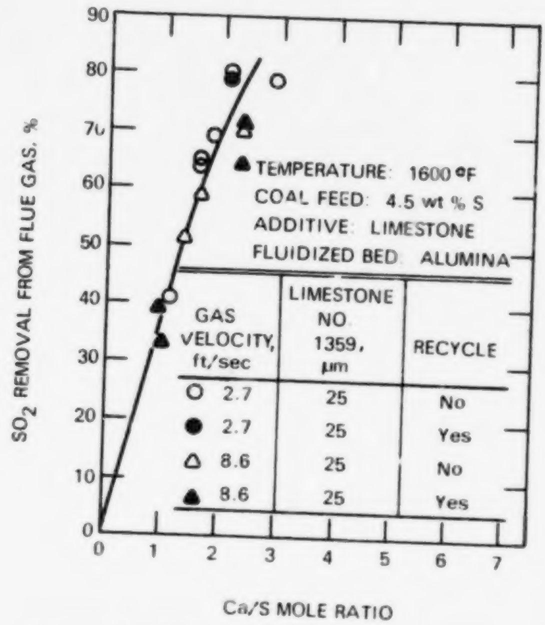


Figure 2 Effect of calcium/sulfur mole ratio on SO$_2$ removal with fine limestone additive.

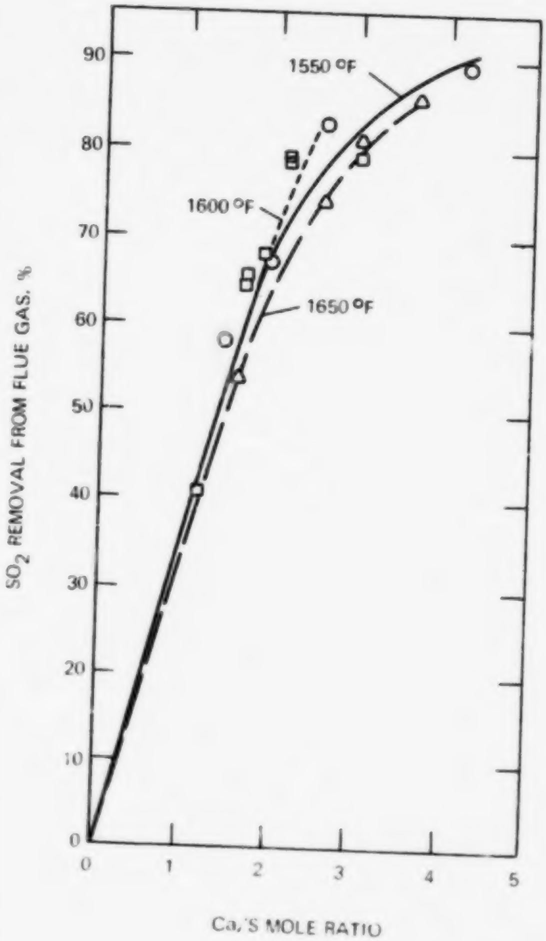


Figure 4. Effect of combustion temperature on SO$_2$ removal with fine limestone additive.

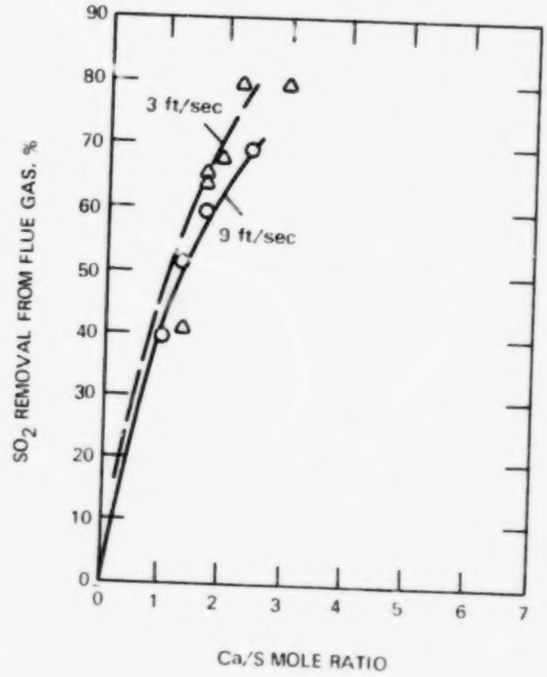


Figure 3. Effect of gas velocity on SO$_2$ removal with fine limestone additive.

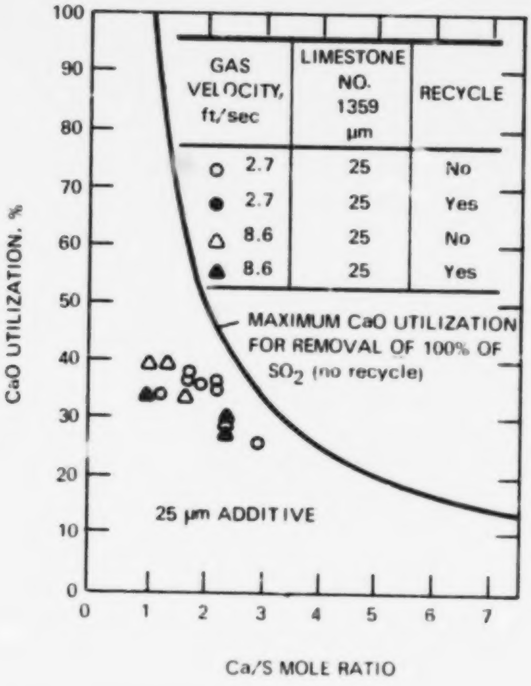


Figure 5. Utilization of CaO as a function of calcium/sulfur mole ratio.

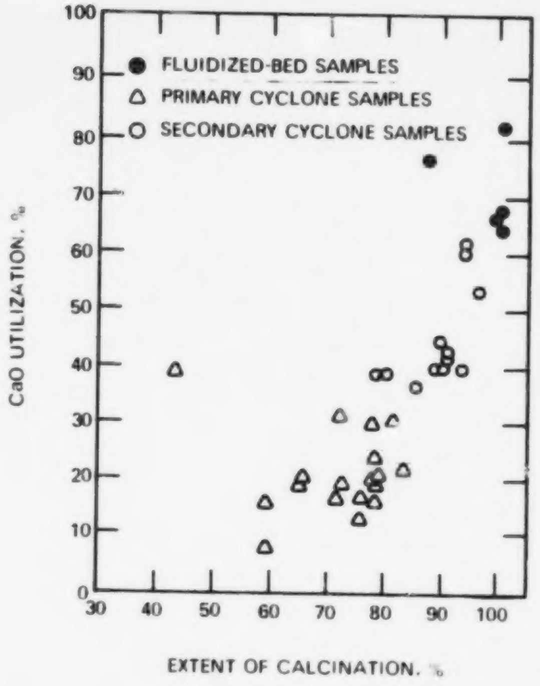


Figure 6. Relationship of CaO utilization to extent of stone calcination.

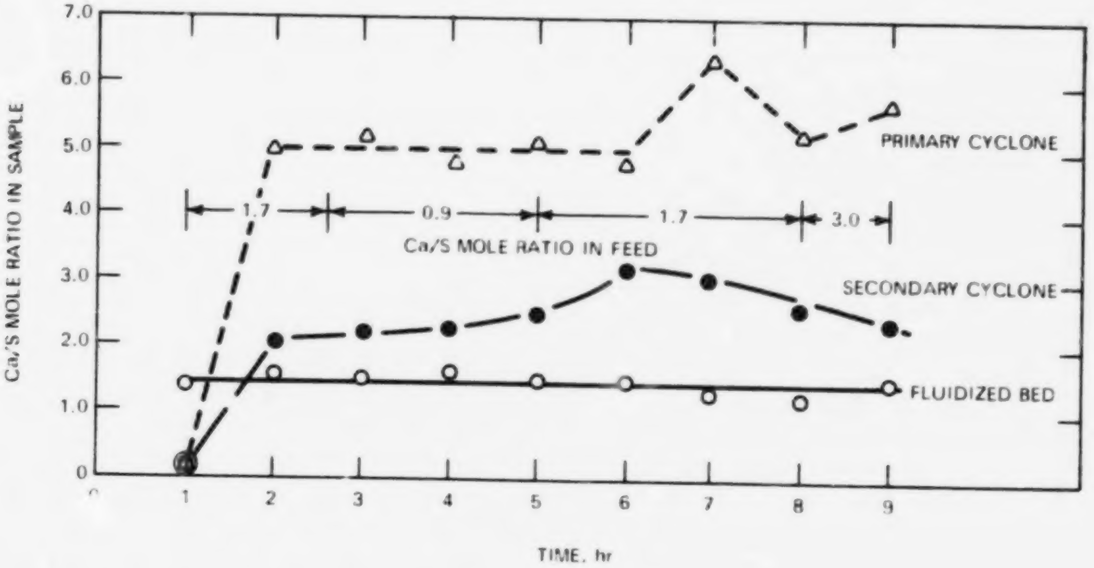


Figure 7. Calcium/sulfur mole ratio in fluidized bed and in elutriated fines during a coal combustion experiment. 25-µm limestone No. 1359.

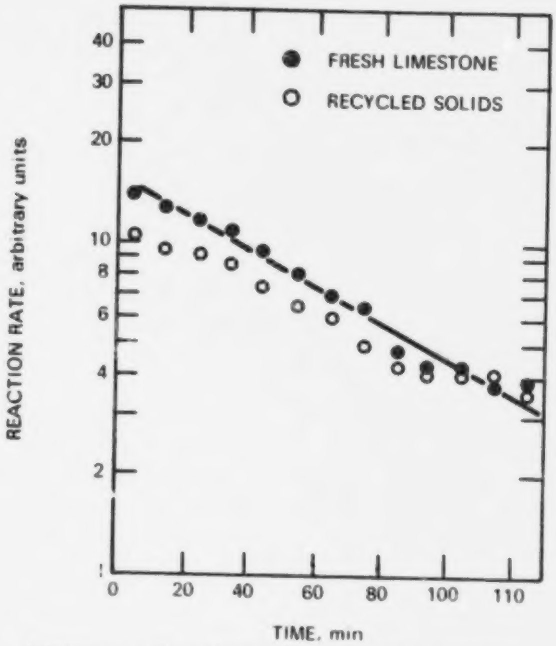


Figure 8. Reaction rates at an SO_2 partial pressure of 3.2 mm Hg for fresh limestone and recycled solids.

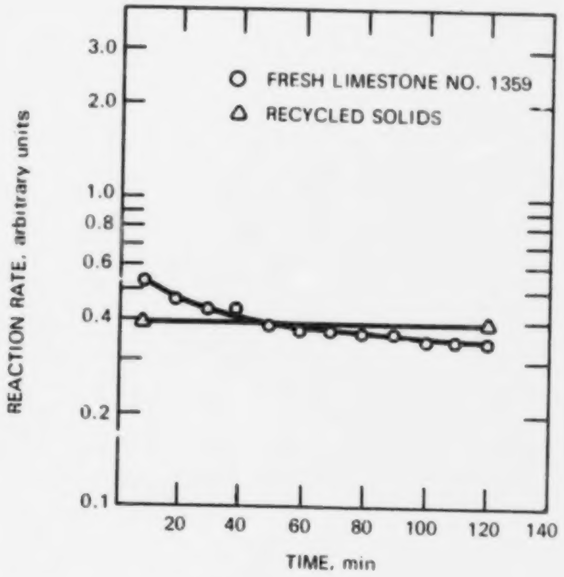


Figure 9. Reaction rates at an SO_2 partial pressure of 0.4 mm Hg for fresh limestone and recycled solids.

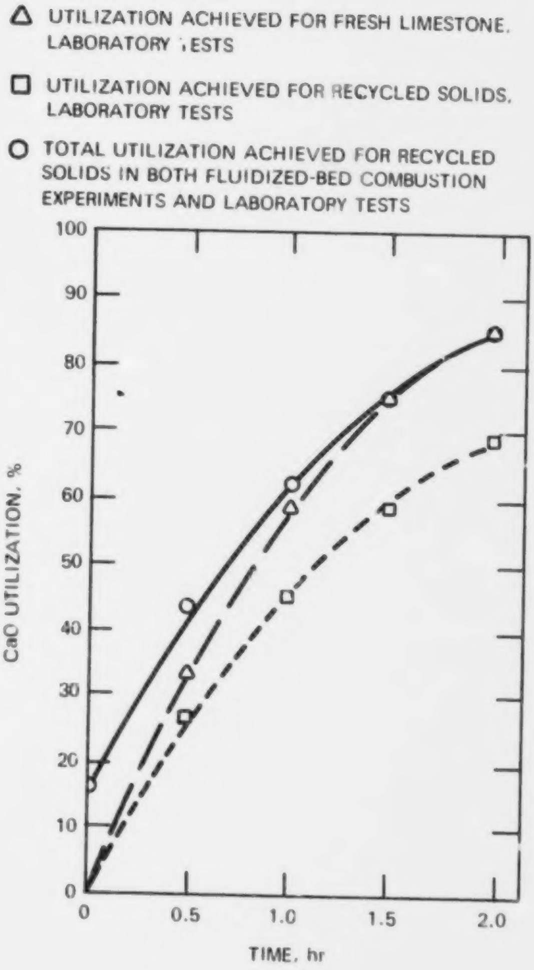
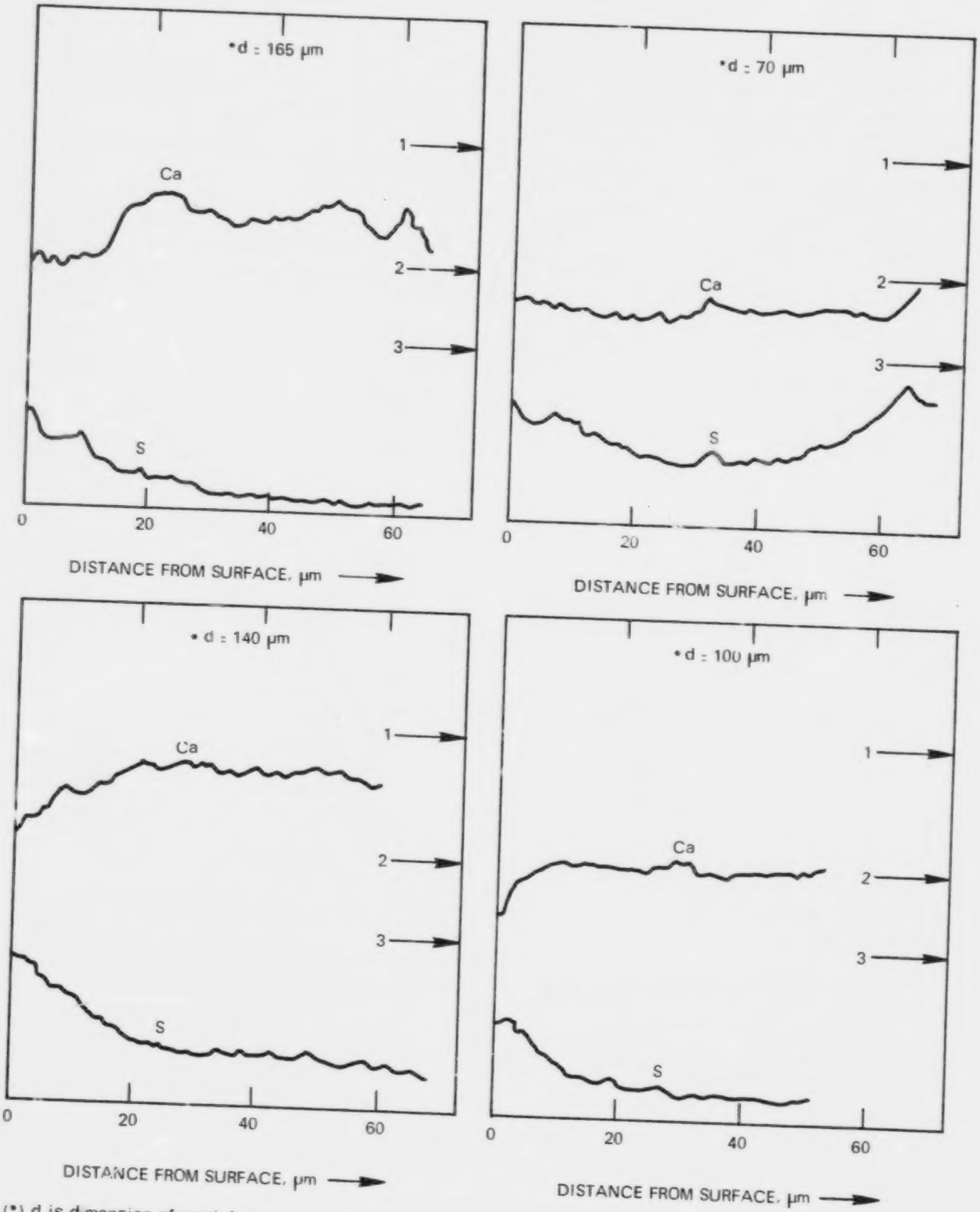


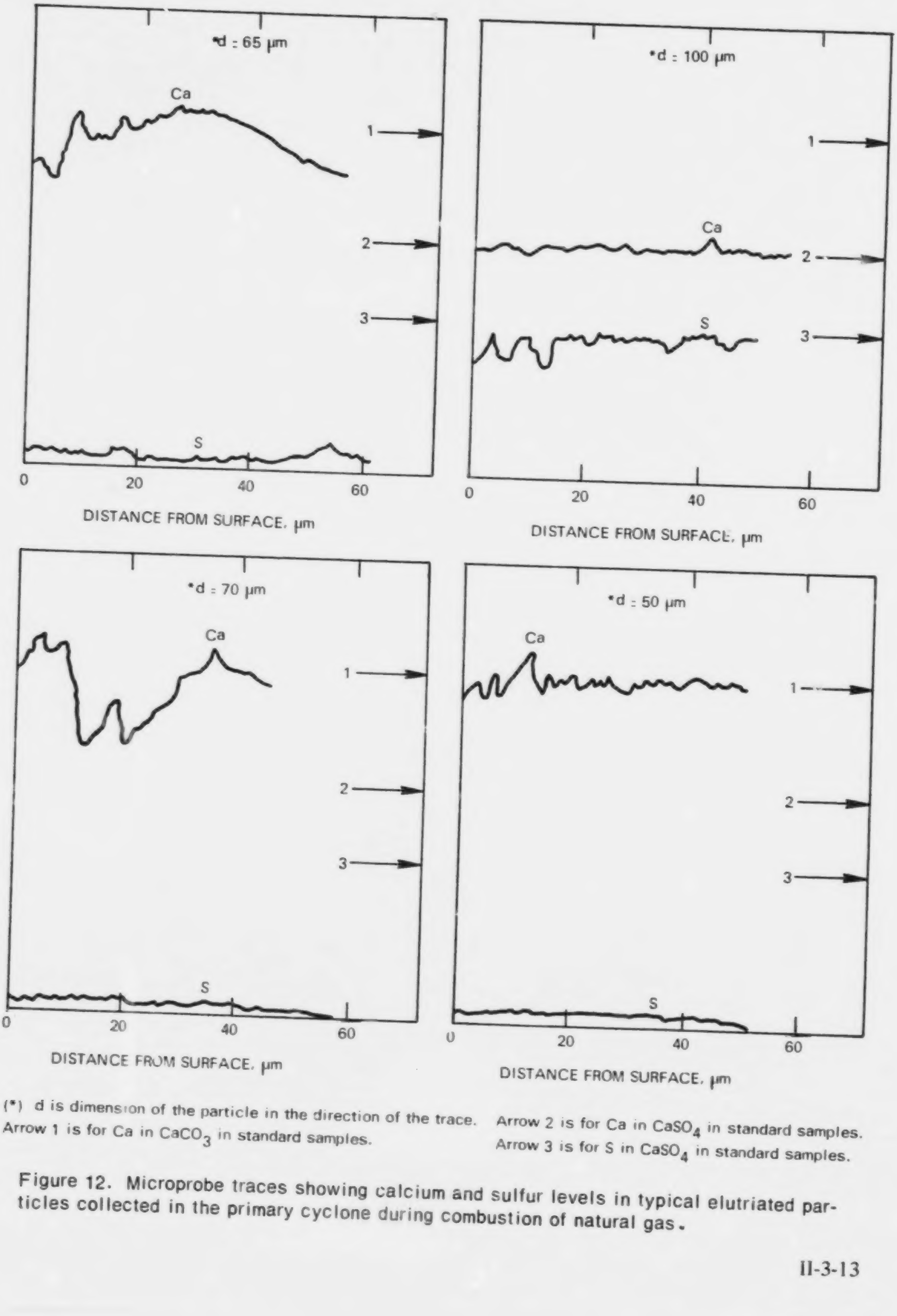
Figure 10. CaO utilization for fresh limestone No. 1359 and for recycled solids.



(*) d is dimension of particle in the direction of the trace.
Arrow 1 is for Ca in $CaCO_3$ in standard samples.

Arrow 2 is for Ca in $CaSO_4$ in standard samples.
Arrow 3 is for S in $CaSO_4$ in standard samples.

Figure 11. Microprobe traces showing calcium and sulfur levels in typical elutriated particles collected in the primary cyclone during combustion of coal.



(*) d is dimension of the particle in the direction of the trace.
Arrow 1 is for Ca in $CaCO_3$ in standard samples.
Arrow 2 is for Ca in $CaSO_4$ in standard samples.
Arrow 3 is for S in $CaSO_4$ in standard samples.

Figure 12. Microprobe traces showing calcium and sulfur levels in typical elutriated particles collected in the primary cyclone during combustion of natural gas.

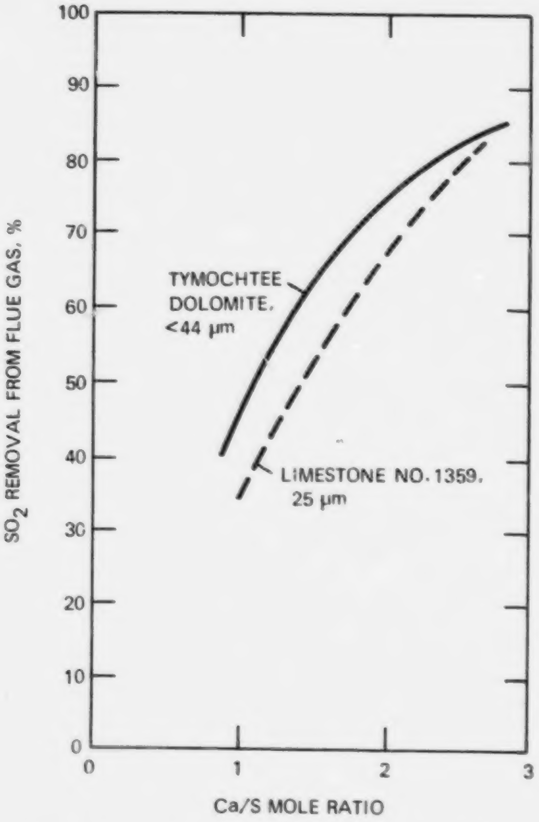


Figure 13. Control of SO$_2$ emission with Tymochtee dolomite No. 1359 limestone.

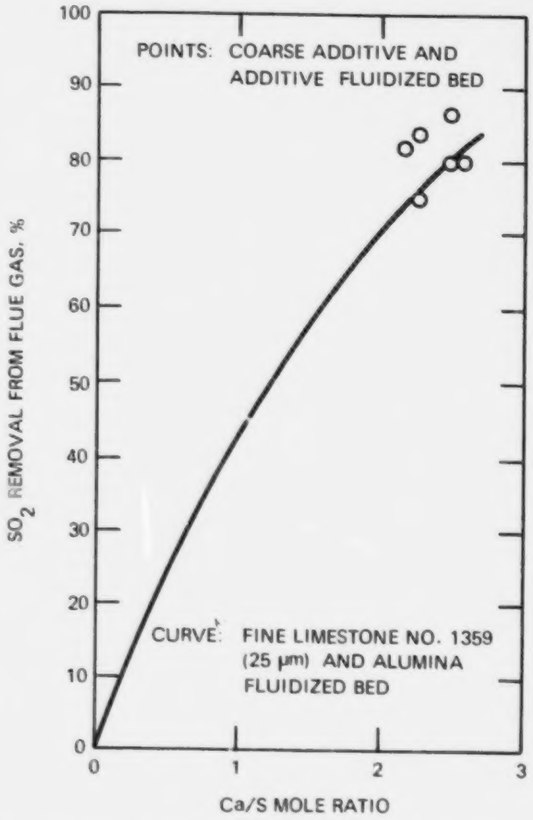


Figure 14. SO$_2$ removal with coarse additive and inert fluidized beds and with fine additive and additive fluidized beds.

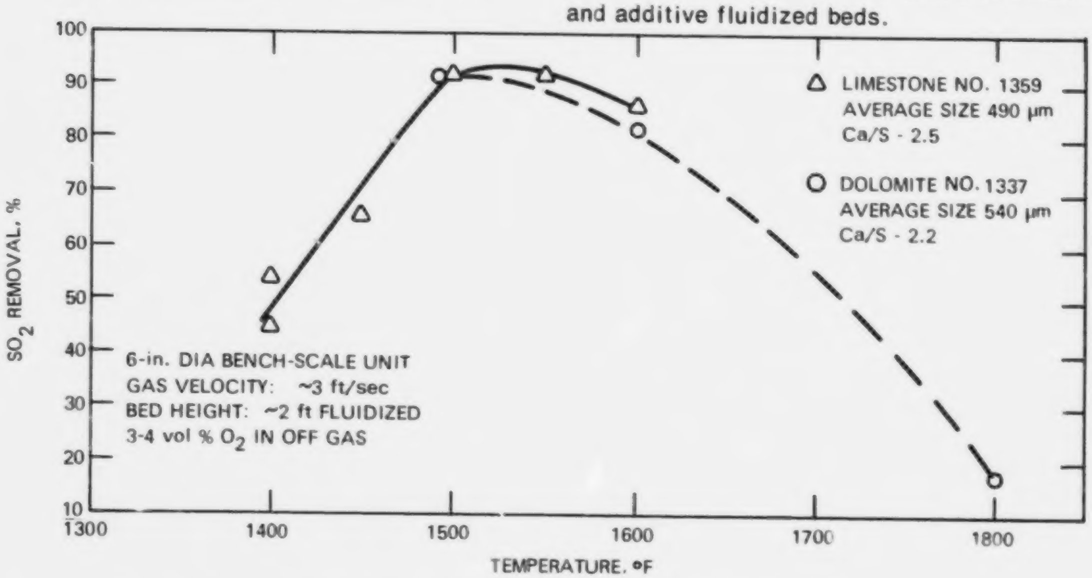


Figure 15. Effect of fluidized-bed combustion temperature on SO$_2$ removal.

4. A REGENERATIVE LIMESTONE PROCESS FOR FLUIDIZED-BED COAL COMBUSTION AND DESULFURIZATION

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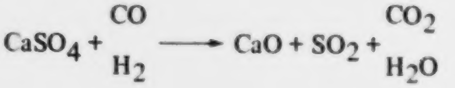
ABSTRACT

The desulfurization efficiency attained on a 3-inch ID fluidized-bed coal combustor operating with a limestone bed is reported as a function of the independent variables of the system. A kinetic model was derived for use in design studies; the reactor length-to-diameter ratio (L/D) was suggested as a scaling parameter in this model. High L/D ratios were found to give relatively poor desulfurization efficiency. It was demonstrated experimentally that the stone maintains a relatively high capacity with repeated cycles of combustion and reductive regeneration. NO_x emissions decreased as a combustion run progressed and the average SO_2 concentration in the batch reactor increased. This indicated a possible interaction between SO_x and NO_x which resulted in lowered NO_x emissions. Small scale experiments using a simulated flue gas showed that SO_2 and NO did react to an appreciable extent in the absence of oxygen. A different mechanism appeared to be operative in the presence of oxygen but a decrease in NO_x was still observed.

INTRODUCTION

Esso Research and Engineering Company is conducting an experimental program for the National Air Pollution Administration (under NAPCA contract CPA 70-19) on a system in which finely ground coal is combusted in a fluidized bed of limestone. The Esso study is a part of NAPCA's overall program to examine fluidized-bed combustion as a possible new boiler (power regeneration) technique. The air pollution control potential of a fluidized-bed combustor is great because of the good gas-solid contacting and, consequently, by appropriate choice of bed material, the high SO_2 removal efficiency which can be attained. Limestone appears to be one of the most promising materials for capture of SO_2. The sulfur in the coal is oxidized to SO_2, which is then captured by the lime as $CaSO_4$. The system proposed by Esso involves transferring the partially sulfated stone from the combustor to a separate regeneration vessel in which the following reaction occurs:



$$\mathrm{CaSO_4} + \begin{matrix} \mathrm{CO} \\ \mathrm{H_2} \end{matrix} \longrightarrow \mathrm{CaO} + \mathrm{SO_2} + \begin{matrix} \mathrm{CO_2} \\ \mathrm{H_2O} \end{matrix} \qquad (1)$$

The regenerated stone (CaO) can then be returned to the combustor for further use. This method of operation naturally reduces the fresh limestone requirements substantially. The off gas from the regeneration unit has a high SO_2 concentration and can be utilized as a feed to a byproduct sulfur or sulfuric acid plant. Figure 1 is a schematic diagram of this process.

Finely ground coal (∿200 μ) is being utilized in the experimental program to facilitate separation of the coal ash from the limestone bed material by entrainment of the coal ash from the bed.

Esso's fluidized-bed combustion program has four primary objectives:

1. Investigate reactor combustion efficiency when feeding finely ground coal.
2. Investigate flue gas desulfurization attainable as a function of the independent variables of the system.
3. Determine the potential for high temperature reductive regeneration of the sulfated bed material.
4. Determine the level of NO_x emissions from the Esso fluidized-bed combustor and investigate possible catalytic reduction of NO_x by limestone.

This presentation is concerned primarily with the air pollution control aspects of the program; i.e., the last three objectives.

EXPERIMENTAL EQUIPMENT

Two fluidized-bed units–a combustor and a regenerator–are being utilized in the Esso experimental study. Operation in these units during regeneration studies is on an intermittent basis: a batch of limestone is charged to the combustor; coal is continuously fed to the combustor until the desired level of limestone sulfation is achieved; the combustor is cooled; and the partially sulfated material from the combustor is transferred to the regenerator. Following regeneration, the solids are returned to the combustor, and another cycle is begun.

Figure 2 is a flow diagram of the Esso fluidized-bed combustor (FBC). The reactor is a 3-inch ID incoloy tube. Four continuous flue gas analyzers are used: NDIR SO_2 and CO analyzers, and polarographic NO_x and O_2 analyzers.

Figure 3 is a flow diagram of the fluidized-bed regeneration unit. The reactor is a 2-inch ID alumina-ceramic tube. An NDIR analyzer is used for continuous measurement of SO_2 in the off gas.

EXPERIMENTAL RESULTS

Range of Variables Examined

Table 1 shows the range of variables examined in the Esso program. Only one coal and one limestone have been utilized in this study.

Table 1. RANGE OF VARIABLES EXAMINED

Variable	Range
Combustor	
Bed temperature	1500-1800°F
Settled bed height	4-16 in.
Superficial gas velocity	2-4 fps
Excess air	3-50%
Average stone particle diameter	460-930 μ
Average coal particle diameter	200 μ
Coal	Bit. (A) (3% S)
Limestone	N-1359
Regenerator	
Bed temperature	2000°F
CO	10 mol %
CO_2/CO	2
Superficial gas velocity	2 fps

Reactor Combustion Efficiency

Table 2 shows the effect of the unit operating conditions upon the reactor combustion efficiency. Perhaps the most unusual effect was on bed height. Increasing the bed height might be expected to cause a longer solid residence time and, consequently, a higher combustion efficiency. The opposite effect was observed in this study; i.e., as bed height was increased, combustion efficiency decreased. This effect is believed to be the result of the slugging nature of the small Esso FBC. This slugging apparently causes a reduced coal particle residence time as bed height is increased. Hence, the increased velocity of the coal particles through the bubble phase in deep beds apparently more than offsets the effect of the additional bed height.

Table 2. EFFECT OF INDEPENDENT VARIABLES ON COMBUSTION EFFICIENCY

Variable	Increasing this variable
Bed temperature (1500-1800°F)	Increases combustion efficiency (91-97%)
Settled bed height (4-16 in.)	Decreases combustion efficiency (94-91%)
Superficial gas velocity (2-4 fps)	Decreases combustion efficiency (97-91%)
Excess air (3-50%)	No effect above 10% excess air
Average stone particle diameter (460-930 μ)	No effect

Desulfurization

The desulfurization reaction was investigated over the range of conditions previously shown in Table 1. Figure 4 shows typical batch desulfurization data obtained. The capacity of the stone at 20 percent SO_2 breakthrough has been chosen as a relative measure of the desulfurization efficiency at a given set of conditions. Table 3 shows the effect of the reactor operating conditions upon the desulfurization efficiency of the Esso FBC. The effect of bed height upon the desulfurization efficiency again apparently reflects the slugging nature of the small Esso fluid bed. Increasing the excess air level decreased the stone capacity. One possible explanation which has been advanced[1] for a similar effect is as follows: at high excess air levels (high O_2 content in the bed), the following reaction sequence can occur:

$$CaO + SO_2 \longrightarrow CaSO_3 \qquad (2)$$

$$CaSO_3 + 1/2\ O_2 \longrightarrow CaSO_4 \qquad (3)$$

At high O_2 levels, reaction (3) occurs quickly and apparently causes a pore blockage, restricting the accessible CaO. However, at low excess air levels, reaction (3) does not proceed as quickly so that less pore blockage by the $CaSO_4$ is experienced.

Table 3. EFFECT OF INDEPENDENT VARIABLES ON STONE CAPACITY

Variable	Increasing this variable
Excess air (3-30%)	Decreases stone capacity[a] (23-12%)
Average stone particle diameter (460-930 μ)	Decreases stone capacity (32-18%)
Superficial gas velocity (2-4 fps)	No effect
Bed temperature (1500-1800°F)	Not effective above 1600°F
Settled bed height (4-16 in.)	Decreases stone capacity (20-7%)

[a] Capacity at 20% SO_2 breakthrough.

Kinetics of Desulfurization

A kinetic model derived in a previous Esso study[2] has been utilized to correlate the desulfurization results from the present work. Figure 5 gives the model assumptions and development. Apparent rate constant k(X) is a function of CaO utilization X.

Figure 6 shows the experimental kinetic data from the Esso FBC program. Constant k(X) is observed to be a function of bed height, as well as CaO utilization. This bed height effect is believed to be a result of poorer gas-solid contacting (because of slugging) as bed height is increased.

A comparison was made between the kinetic data obtained in this study and data obtained in a previous Esso study[2] in which a simulated flue gas was desulfurized. Figure 7 shows that the two sets of kinetic data agree closely. The implication from this comparison is that a large fraction of the SO_2 generated during coal combustion in the Esso FBC is released near the reactor inlet.

In this study, the data shown in Figure 6 has been replotted at constant values of the L/D (aspect) ratio. We believe that by maintaining a constant L/D ratio in scale-up, similar contacting efficiencies can be expected. Figures 8 and 9 show that decreasing the L/D ratio at a constant set of other operating conditions increases the percentage of available CaO which can be utilized in achieving a given SO_2 removal. Hence, it appears that high values of the L/D ratio will give conservative estimates of the performance of large scale equipment, in which the L/D ratio is less than unity. For design purposes, the desulfurization data shown in Figure 8 at an L/D of 3.7 should provide a conservative estimate of commercial reactor performance.

Stone Regeneration

The partially sulfated stone was regenerated by reduction at 2000°F. A 2/1 volumetric ratio of CO_2/CO was maintained in the reducing gas to minimize sulfide formation. The reducing gas contained 10 mol percent CO. Equation (1) is the primary reaction occurring in the regenerator.

Figure 10 shows the SO_2 concentration in the regenerator off gas. The dashed line indicates the SO_2 concentration that would be attained at thermodynamic equilibrium. The occasional low values of SO_2 concentration in the off gas are believed to represent poor gas-solid contacting in the regenerator rather than any kinetic limitations. The system can be expected to attain thermodynamic equilibrium in a commercial system.

Cycling of the bed material between combustion and regeneration gave the stone capacity data shown in Figure 11. Initially the capacity of the regenerated stone was higher than that of the fresh stone. This phenomenon is possibly a result of a more favorably crystal lattice, as compared to the fresh stone, formed on eliminating SO_3 from the sulfated material. Sorbent activity slowly decreased on repeated regeneration. Also shown in Figure 11 is cyclic stone capacity data obtained in a previous Esso study[2] using a simulated flue gas for SO_2 sorption.

NO_x Emissions

Figure 12 is a typical record of NO_x emissions from the Esso FBC. As a run progressed, and the SO_2 level in the bed and flue gas increased, the NO_x emissions decreased. The indication is that there is some interaction between the SO_x and NO_x which causes the NO_x emissions to decrease.

In order to investigate what type of interaction might be occurring between NO_x and SO_2, a series of experiments (using a simulated gas with varying concentrations of NO and SO_2) was conducted in the 2-inch regeneration reactor. Table 4 shows the results of this exploratory series of experiments.

Table 4. NO_x-SO_2 INTERACTION IN A FLUIDIZED BED[a] OF LIMESTONE

Bed Material	NO_x Conc., ppm		SO_2 Conc., ppm	
	Before SO_2 Intro.	After SO_2 Intro.	Inlet	Outlet
	Transport Gas - N_2			
Gas phase	900	900	1290	1290
Partially sulfated stone from combustor	840 830	440 180	785 1510	300 480
Alundum	820	820	1000	1000
	Transport Gas Air			
Gas phase	800	760	1290	1180
Partially sulfated stone from combustor	710	710	785	0
Alundum	620	380	1090	1090

[a] Bed temperature–1630°F.

The indication is that the NO and SO_2 can react in a partially sulfated bed in the absence of O_2; no reaction occurs over an inert bed in the absence of O_2. One possible reaction mechanism which can be occurring is:

$$CaO + SO_2 \longrightarrow CaSO_3 \quad (4)$$

$$CaSO_3 + 2NO \longrightarrow CaSO_3 \cdot (NO)_2 \quad (5)$$

$$CaSO_3 \cdot (NO)_2 \longrightarrow CaSO_4 + N_2 + 1/2\ O_2 \quad (6)$$

However, in the presence of O_2, reaction occurs between NO_x and SO_2 even over an inert bed. Although no reaction mechanism has been identified, the oxidized species, NO_2 and SO_3, could possibly be involved.

Further studies will be made of the exact nature of SO_x - NO_x interaction in the presence of O_2. Methods of utilizing this interaction to effect low NO_x emissions from a commercial reactor will also be investigated.

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2. Skopp, A. et al. Fluid Bed Studies of the Limestone Based Flue Gas Desulfurization Process. Final Report under NAPCA Contract PH 86-67-130, August 1969.

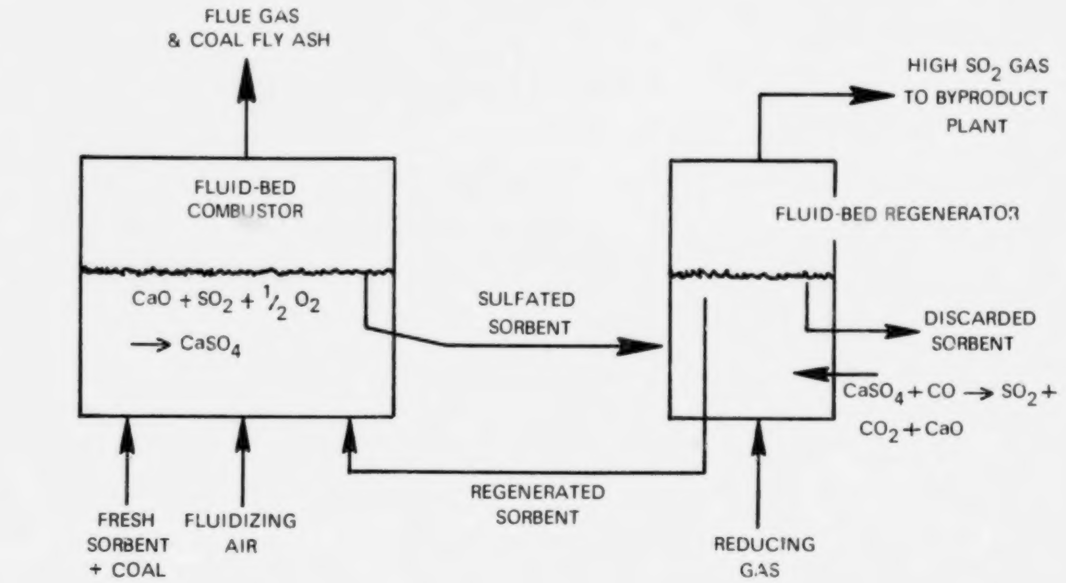

FLUE GAS
& COAL FLY ASH
FLUID-BED
COMBUSTOR
CaO + SO2 + 1/2 O2
→ CaSO4
SULFATED
SORBENT
FLUID-BED REGENERATOR
HIGH SO2 GAS
TO BYPRODUCT
PLANT
DISCARDED
SORBENT
CaSO4 + CO → SO2 +
CO2 + CaO
REGENERATED
SORBENT
FRESH
SORBENT
+ COAL
FLUIDIZING
AIR
REDUCING
GAS

Figure 1. Regenerative limestone system proposed by Esso Research.

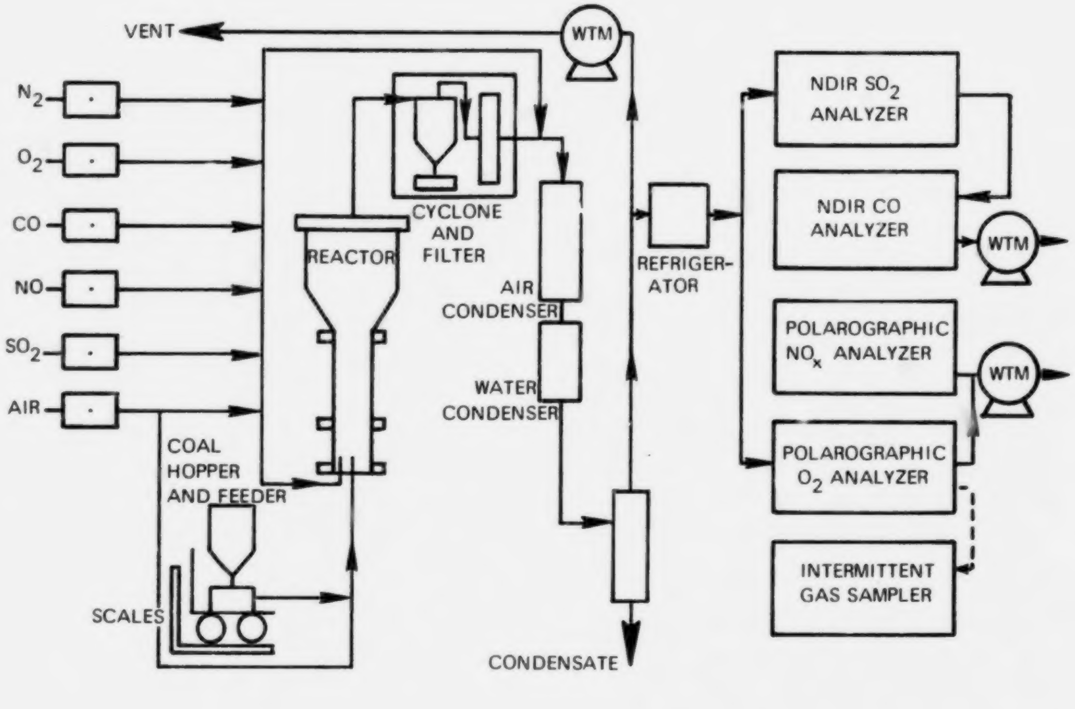

VENT
WTM
N2
O2
CO
NO
SO2
AIR
CYCLONE
AND
FILTER
REACTOR
AIR
CONDENSER
WATER
CONDENSER
COAL
HOPPER
AND FEEDER
SCALES
CONDENSATE
REFRIGER-
ATOR
NDIR SO2
ANALYZER
NDIR CO
ANALYZER
WTM
POLAROGRAPHIC
NOx ANALYZER
WTM
POLAROGRAPHIC
O2 ANALYZER
INTERMITTENT
GAS SAMPLER

Figure 2. Esso fluidized-bed combustion unit.

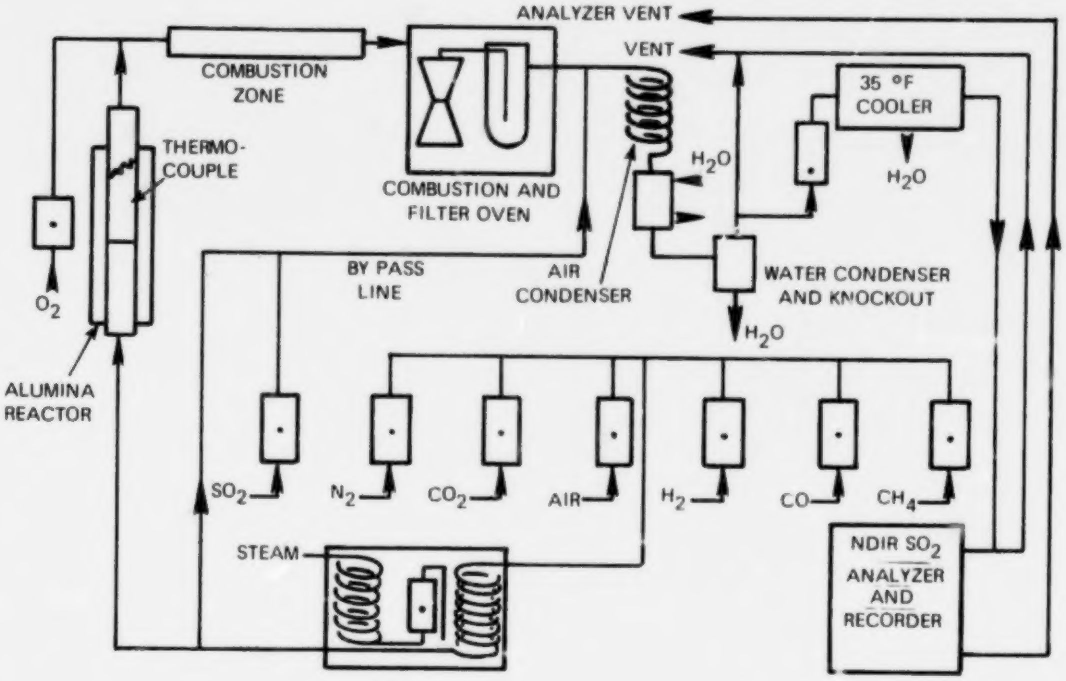

ANALYZER VENT
VENT
COMBUSTION ZONE
THERMO-COUPLE
COMBUSTION AND FILTER OVEN
H_2O
35 °F COOLER
H_2O
O_2
BY PASS LINE
AIR CONDENSER
WATER CONDENSER AND KNOCKOUT
H_2O
ALUMINA REACTOR
SO_2
N_2
CO_2
AIR
H_2
CO
CH_4
STEAM
NDIR SO_2 ANALYZER AND RECORDER

Figure 3. Flow diagram for the fluidized-bed regeneration unit.

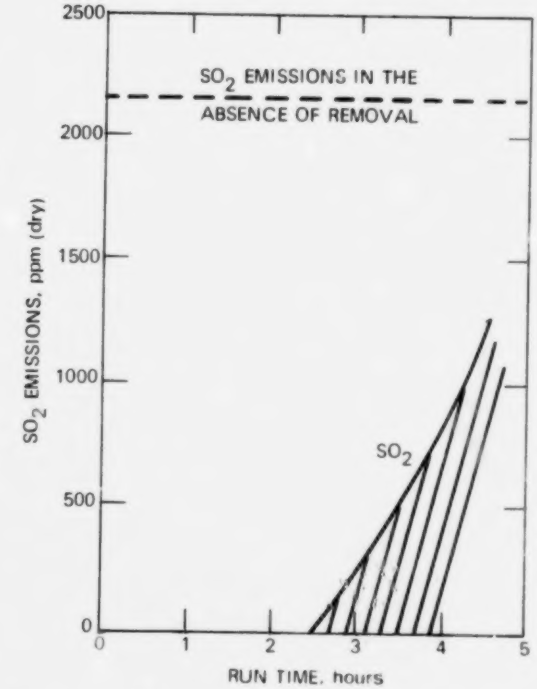
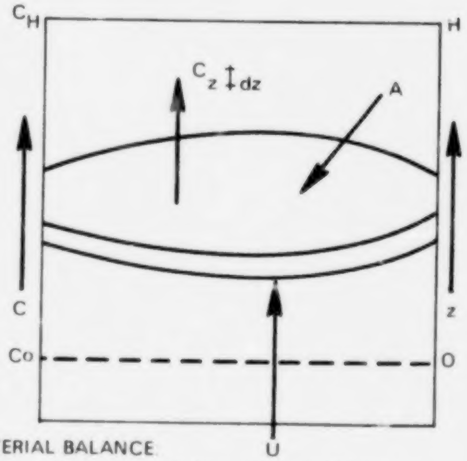


Figure 4. Typical batch desulfurization data.


C_H
H
$C_z \updownarrow dz$
A
C
z
Co
O
U

MATERIAL BALANCE

SO_2 REMOVED FROM GAS = SO_2 ABSORBED BY SORBENT

YIELDS: $\ln (C_H/C_o) = -k(X) \cdot Ho/U$

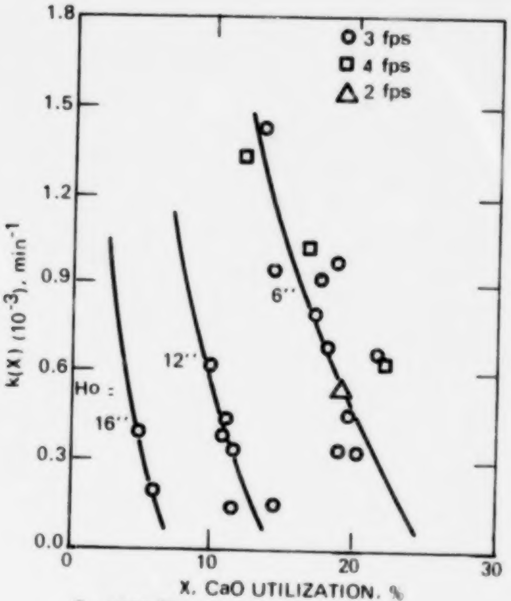
MODEL ASSUMPTIONS

COMPLETE MIXING OF SOLIDS

PLUG FLOW OF GAS

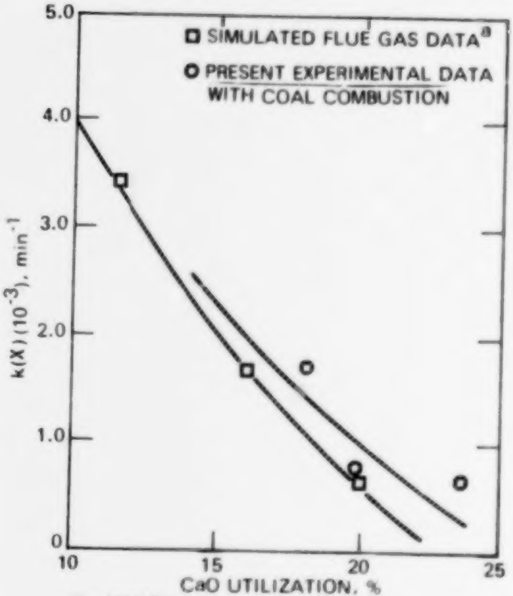
ALL SO_2 RELEASED AT REACTOR INLET

Figure 5. Esso kinetic model development.



T : 1600 °F
EXCESS AIR ~10%
(dp) COAL : 200 μ
(dp) STONE : 930 μ

Figure 6. Kinetic constants from Esso data.

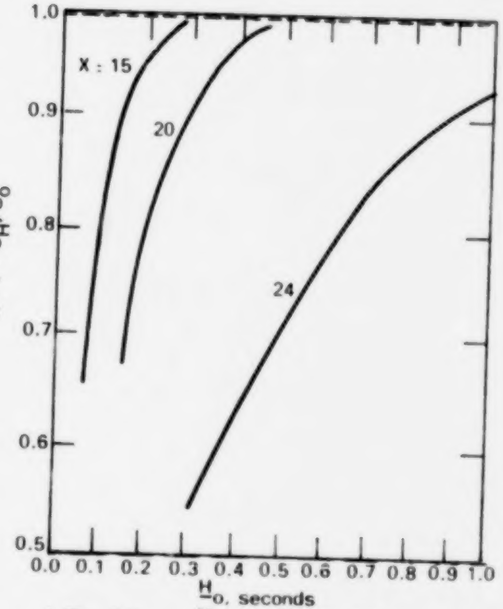


T : 1600 °F
U : 3-4 FPS
H_o : 4 in.

(dp) COAL : 200 μ
EXCESS AIR ~10%
(dp) STONE : 930 μ

[a] Data from reference 2.

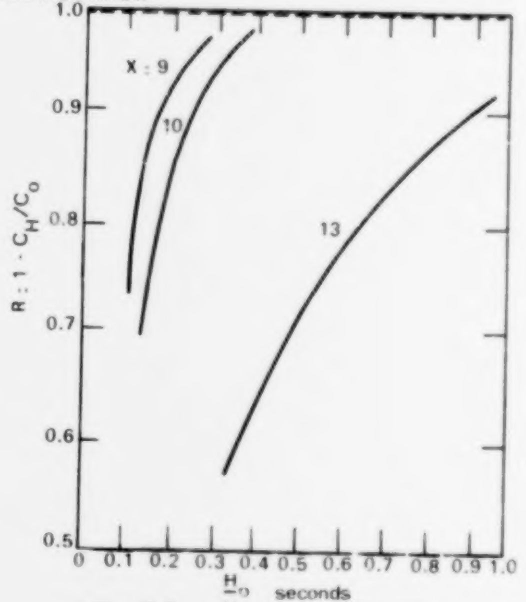
Figure 7. SO_2 formation occurs primarily near reactor inlet.



L/D : 3.7
EXCESS AIR ~10%

(dp) STONE : 930 μ
(dp) COAL : 200 μ

Figure 8. Kinetic results at L/D = 3.7.



L/D : 10.3
EXCESS AIR ~10%

(dp) STONE : 930 μ
(dp) COAL : 200 μ

Figure 9. High L/D gives low stone utilizations.

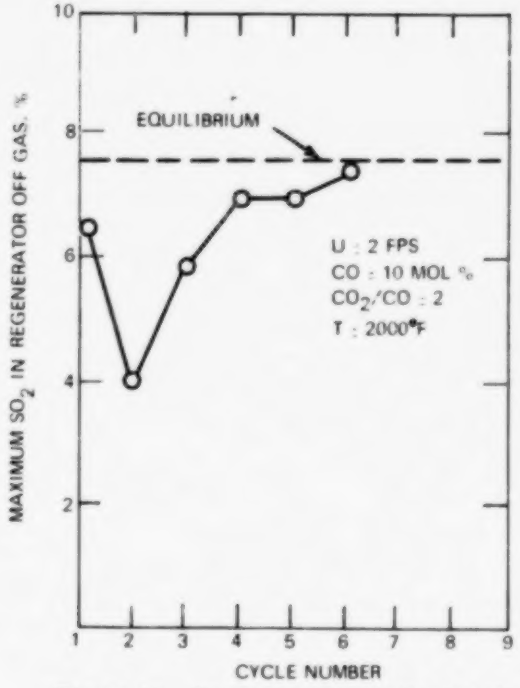


Figure 10. Regeneration reaction attains equilibrium.

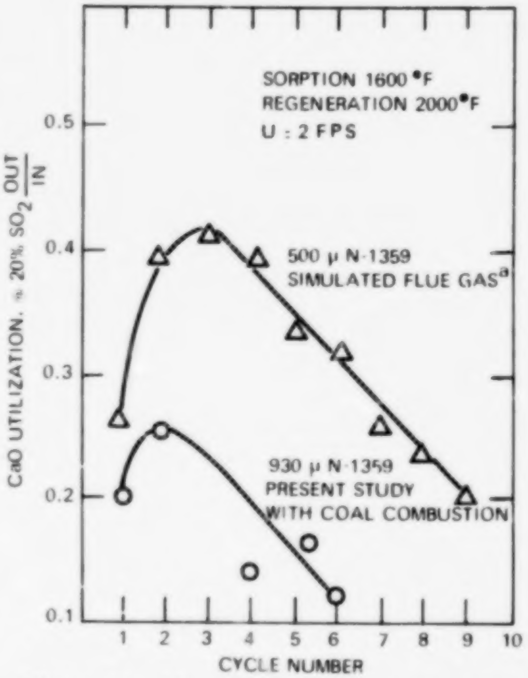


Figure 11. Absorption reactivity upon cycling.

[a]Data from reference 2.

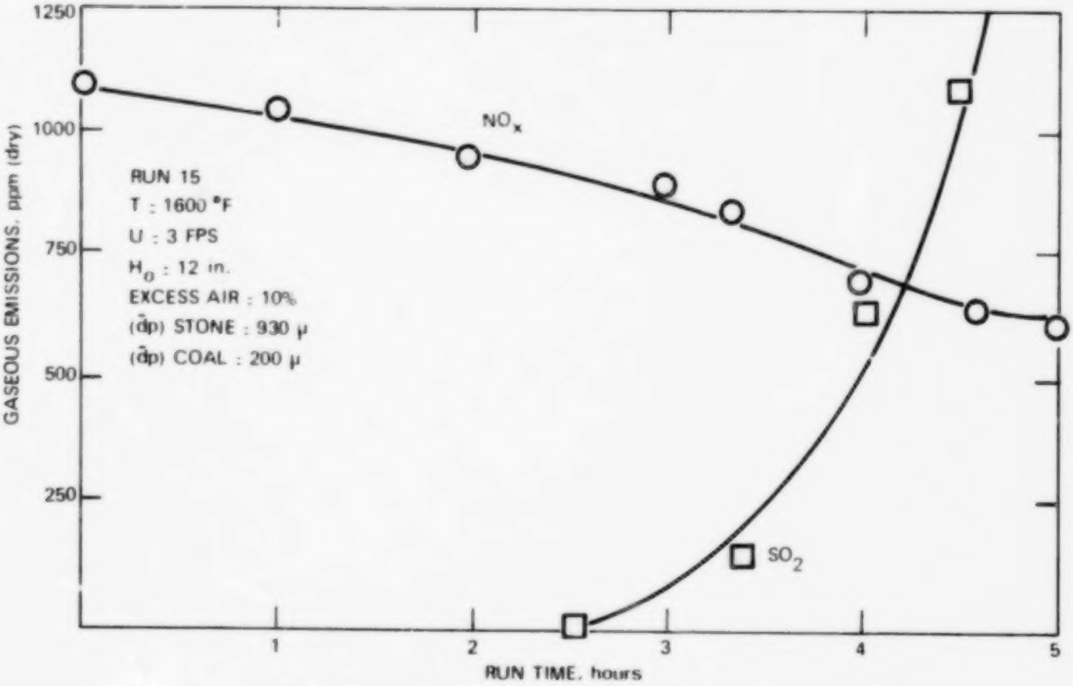


Figure 12. Typical NO_x emissions with limestone bed.

5. COAL-BASED SULFUR RECOVERY CYCLE IN FLUIDIZED-LIME-BED COMBUSTION

G. P. CURRAN, C. E. FINK, AND EVERETT GORIN

Consolidation Coal Company

ABSTRACT

Sulfated dolomite was produced by the combustion of coal in a fluidized bed of dolomite continuously fed with raw dolomite. A brief experimental study is reported here of a two-stage process for regeneration of the $CaSO_4$; i.e., its conversion to CaO, by use of coal as a reductant. The composition of the off gases from this process is controlled so that just enough $CO + H_2$ is available to reduce the SO_2 to elemental sulfur.

The results of a computer study on management of the off gas to obtain maximum yield of sulfur are also presented.

INTRODUCTION

A previous paper[1] from these laboratories presented data on the efficiency of removal of sulfur in the combustion of coal within a fluidized bed of dolomite. The sulfur is fixed on the dolomite in the form of calcium sulfate. Regeneration of the calcium sulfate to reform calcium oxide also was investigated to obtain data on acceptor life and dolomite makeup requirements.

The regeneration in the previous study was conducted by the use of partial combustion with air of CO as a fuel gas. The essential regeneration reaction is endothermic and may be written,

$$CaSO_4 + CO = CaO + CO_2 + SO_2 \qquad (1)$$
$$\Delta H = +53{,}720 \text{ cal/g mol at } 25°C.$$

Hence, the need for partial combustion to supply heat.

Although the procedure was successful as a laboratory expedient, it would be much more practical in a commercial situation to use coal as a reductant and fuel; i.e.,

$$CaSO_4 + C + 1/2\,O_2 = CaO + SO_2 + CO_2 \qquad (2)$$
$$\Delta H = +27{,}520 \text{ cal/g mol}$$

Attempts to use char from high-sulfur Pittsburgh seam coals for this purpose were unsuccessful. It was found that under reducing conditions and at the high temperatures (>1900°F) required to effect reaction (2), ash fusion was an insurmountable problem.

Accordingly, to avoid this problem a two-stage regeneration process was devised, using coal as fuel. This paper describes the process and presents preliminary experimental data which demonstrates its feasibility.

This paper also discusses the thermodynamics of recovery of elemental sulfur from the regeneration off gases, and outlines the net savings in dolomite and energy requirements by use of the process.

PROCESS DESCRIPTION

Figure 1 is a schematic sketch of the overall process. In the first stage, $CaSO_4$ is reduced to CaS at a relatively low temperature (about 1850°F). In principle, $CaSO_4$ can be reduced to CaS by a series of reactions which can be expressed as the overall reaction,

$$CaSO_4 + 2\,C = CaS + 2\,CO_2 \qquad (3)$$

$$\Delta H = +42{,}440 \text{ cal/g mol}$$

In practice, however, additional carbon must be burned to produce CO. The overall reaction then can be expressed as,

$$CaSO_4 + \left[\frac{X}{1\text{-}A}\right] C + \left[\frac{X\,(2\text{-}A) - 4\,(1\text{-}A)}{2\,(1\text{-}A)}\right] O_2 \qquad (4)$$

$$= CaS + X\,CO_2 + \frac{AX}{(1\text{-}A)}\,CO$$

where: $\frac{X}{1\text{-}A}$ = mol ratio of $C/CaSO_4$ required for process heat balance, and
A = $CO/\,(CO + CO_2)$, mol ratio in exit gas to satisfy the reaction kinetics.

Another feature of the first-stage process is that it is so conducted that sufficient reducing gas is produced to reduce the SO_2 formed in the second stage to sulfur by reactions such as,

$$SO_2 + 2\,CO = 1/2\,S_2 + 2\,CO_2 \qquad (5)$$

$$\Delta H = -48{,}900 \text{ cal/g mol}$$

Thus, $AX/(1\text{-}A) = 2$ and reaction (4) becomes

$$CaSO_4 + 3.890\,C + 0.890\,O_2 \qquad (4a)$$

$$= CaS + 1.890\,CO_2 + 2\,CO$$

$$\Delta H = O$$

The high CO/CO_2 ratio in reaction (4a) ensures that the $CaSO_4$ reduction will occur rapidly. With the use of coal, combustion of its hydrogen content produces additional heat, which is needed to preheat the incoming coal and air. With respect to the coal-air reaction, the first stage is a fluidized-gas producer in which ash slagging will not occur at nominal bed temperatures of 1850°F because the combustion takes place in the presence of heat sink created by the endothermic reduction of $CaSO_4$ to CaS – reaction (3).

In the second stage, the acceptor, now in the form of CaS, is contacted with air in a fluidized bed at temperatures in the range of 1900-1950°F. No external fuel is used. The sulfur is rejected by a series of reactions which may be expressed as the overall reaction,

$$CaS + 3/2\,O_2 = CaO + SO_2 \qquad (6)$$

$$\Delta H = -108{,}970 \text{ cal/g mol}$$

A portion of the incoming CaS first is oxidized to $CaSO_4$ via

$$CaS + 2\,O_2 = CaSO_4 \qquad (7)$$

Then the $CaSO_4$ reacts with the residual CaS via,

$$3\,CaSO_4 + CaS = 4\,CaO + 4\,SO_2 \qquad (8)$$

to give CaO and SO_2. At a nominal pressure of 1 atm, the incoming air must be diluted with recycled tail gas from the sulfur recovery section in order to provide a ΔP SO_2 driving force for reaction (8). A mol ratio of about

1/1 recycle-gas/air is required. The exothermic reaction provides the preheat duty for the incoming air and recycled gas.

The combined off gases from both stages are treated in the sulfur recovery section where the SO_2 produced in the second stage is reduced to elemental sulfur by CO (and H_2 formed by the water-gas shift reaction) produced in the first stage. Since both $CaSO_4$ and CaS are present in the first stage, some SO_2 inevitably will be formed by reaction (8). In the presence of CO and H_2, this SO_2 will be reduced to H_2S, COS, and S_2 with the last material predominating. This situation is not detrimental, since all sulfur compounds are nearly completely converted to elemental sulfur in the recovery section.

The purpose of the work reported here was to study briefly the salient features of the two-stage regeneration process and the severity of the possible processing problems outlined below.

First Stage

1. At the very low air/fuel ratios required, the incoming raw coal may form coke.
2. To demonstrate freedom from ash slagging at reducing conditions using Eastern steam coal.
3. To determine the operating temperature and coal/$CaSO_4$/air ratios required to generate sufficient CO to satisfy reaction (4a).
4. To determine whether deposits form.

Second Stage

1. To determine the O_2/CaS ratio required for optimum rejection of sulfur as SO_2, as a function of temperature and ΔP_{SO_2} driving forces.
2. Previous experience with the CO_2 acceptor process, in which CaS is converted to CaO and SO_2 in the regenerator under conditions similar to those of the second stage, showed that when CaS and $CaSO_4$ exist simultaneously in the acceptor, a transient liquid formed which led to massive deposits and/or cementing together of the acceptor particles. In this work, it was vital to determine the nature and extent of deposit formation.

EXPERIMENTAL

Generally, the equipment and experimental procedure were the same as reported previously.[1] Figure 2 is a schematic diagram of the equipment.

Briefly, the reactor had a 4-inch I.D. and was made from Type 316 stainless steel. The fluidized-bed height was controlled at 36 inches by an overflow weir. The original reactor was modified by replacing the perforated disc, baffle, and plenum chamber at the bottom with a cone having an included angle of 40 degrees. All solids were fed to the apex of the cone. The same reactor was used for all steps of the process, including coal preoxidation; i.e., no attempt was made to operate all the steps simultaneously.

After reaching the programmed flows and bed temperature, solids feeding was continued nominally for three bed inventory changes and the data was recorded over a 1-hour balance period. By adjusting the electrical power input to each of the three sections of the reactor heater, the maximum temperature gradient across the fluidized bed was held to within 20°F.

The data workup was straightforward, being based on measured values for the input and output streams, the dry exit gas rates, product gas analyses, and the acceptor compositions as determined by a special assay for the CaS and $CaSO_4$ content. In the first-stage runs, considerable sulfur was rejected to the gas through reaction (8). At the conditions used, most of the SO_2 was reduced to S_2 which condensed as fog in the gas recovery system and escaped with the product gas. The amount of elemental sulfur formed was obtained by a forced sulfur balance.

The acceptor was the same Tymochtee dolomite (Western Ohio), sized to 16 x 28 mesh, used in the previous work.

RESULTS AND DISCUSSION

Preoxidation of Coal Feed

All work reported here was carried out with Ireland Mine coal, a high-sulfur, highly caking Pittsburgh seam coal. Early work on feeding raw coal to stage 1 showed that this was not practical due to a small amount of coke formation. Accordingly, it was found necessary to preoxidize the feed coal to reduce its caking propensity.

A level of preoxidation of 5.3 wt percent (defined as pounds of oxygen reacted with 100 pounds MF coal) was found sufficient to prevent coke formation. The preoxidation was carried out by continuous feed of raw coal (28 x 100 mesh) to a fluidized bed at 700°F. A mixture of air and nitrogen was used as fluidizing gas. The composition of the preoxidized coal is:

Hydrogen	4.52, wt percent, MF basis
Carbon	72.44
Nitrogen	1.29
Oxygen (by diff.)	6.71
Sulfur	4.34
Ash	10.70

Gross Btu, MF basis, by Dulong formula, 12,995 Btu/lb.

Stage 1 Runs

After preliminary runs were made to determine the extent of preoxidation required, a series of runs were made with the above preoxidized coal. Five runs were made with fresh sulfated acceptor at 1825 and 1875°F and at various input acceptor, coal and air rates. Another run, to help determine any kinetics effects of acceptor activity loss on the rate of $CaSO_4$ reduction, was made with an acceptor exposed previously to seven combustion-regeneration cycles. Run conditions and results are shown in Table 1.

The objective of these runs was to determine conditions under which three simultaneous conditions could be satisfied: nearly complete reduction to CaS, a ratio of $CO + H_2/CaS = 2$ in the product, and process heat balance. For each run, a complete heat balance was calculated with the results shown in the last row of Table 1. The heat balance was based on air and preoxidized coal fed to the process at 100 and 700°F, respectively.

Comparisons of Runs A7-A7A and A10-A10A show that, by increasing the temperature, the desired ratio, $(CO + H_2)$/(total S in acceptor product), can be achieved easily. Substantially complete reduction of $CaSO_4$ occurred in all the fresh acceptor runs, showing that the reduction reactions are rapid at temperatures of 1825°F and above, even with $(CO + H_2)$ concentrations as low as 9.9 percent in the exit gas, as in Run A8. The acceptor retention times in these runs ranged from 0.8 to 1.1 hours. In run A9, made with deactivated acceptor, the somewhat lower level of $CaSO_4$ reduction may have been caused by the decreased retention time brought about by the increased acceptor feed rate which was used in order to keep the $CaSO_4$ input roughly comparable with that in the other runs. Thus, deactivation has no

Table 1. CONDITIONS AND RESULTS FOR FIRST-STAGE RUNS[a]

Run Number	A7	A7A	A8	A10	A10A	A9
Temperature, °F	1825	1875	1825	1825	1875	1825
	b	b	b	b	b	c
Acceptor Feed						
Feed rate, lb/hr	9.44	9.44	11.26	11.01	9.44	23.72
Comp., mol & of total Ca						
CaO	10.50	10.50	10.50	10.50	10.50	65.50
CaS	0	0	0	0	0	0
$CaSO_4$	89.50	89.50	89.50	89.50	89.50	34.50
Coal Feed Rate, lb/hr	4.79	4.79	4.79	4.93	4.93	4.22
Inlet Gas, SCFH						
Air	152	152	152	125	125	139
N_2 purges	3.6	3.6	3.6	3.6	3.6	3.6
Exit Gas Rate, SCFH Dry Gas	231	235	232	207	203	218
Exit Gas Comp. at Top of Bed						
H_2O, mol %	12.72	12.06	13.80	15.07	13.76	13.61
H_2	3.88	4.65	2.71	3.98	5.88	2.20
CH_4	0.48	0.20	0.46	0.55	0.52	0.43
CO	9.79	12.30	7.21	9.29	13.02	6.58
CO_2	18.81	17.55	21.51	20.61	16.74	23.86
S_2	1.03	1.05	0.96	1.06	1.18	0.81
H_2S	0.29	0.34	0.25	0.26	0.19	0.10
COS	0.03	0.03	0.03	0.02	0.03	0.02
N_2 (diff.)	52.97	51.82	53.07	49.16	48.68	52.39
Product Acceptor						
Rate, lb/hr	6.27	6.25	7.54	7.35	6.27	20.08
Comp., mol % of total Ca						
CaO	29.8	31.3	25.3	27.0	29.8	69.4
CaS	69.6	68.1	73.8	72.2	69.6	28.6
$CaSO_4$	0.6	0.6	0.9	0.8	0.6	2.0
Unreacted Char, lb/hr	1.99	1.88	1.99	2.31	2.33	1.52
Outlet Fluidizing Velocity, ft/sec	2.19	2.27	2.20	1.96	1.96	2.06
Bed Density, lb/ft^3	24.6	23.9	24.5	27.0	27.0	30.7
Bed Weight, lb	6.15	5.98	6.13	6.75	6.75	7.68
Solids Retention Time, hr	0.98	0.96	0.81	0.92	1.08	0.38
% Carbon Burnout $\frac{\text{Lb C Gasified}}{\text{100 Lb C Fed}}$	60.3	63.3	60.7	54.8	53.5	68.1
% S in Acceptor Rejected to Gas	21.5	23.3	16.5	18.4	21.6	10.6
% Reduction, $\frac{CaS}{CaS + CaSO_4}$, Mol Ratio	99.2	99.1	98.8	99.0	99.2	93.5
Mol Ratio, $\frac{CO + H_2}{CaS + CaSO_4}$	2.20	2.84	1.26	1.57	2.67	0.94
Process Heat Balance, Btu Absorbed/lb Mol $CaSO_4$ Fed	-67,700	-48,400	-27,400	-4840	+7800	-19,200

[a] Fuel was 5.3% preoxidized Ireland Mine coal, at an 8-psig system pressure.

[b] Fresh sulfated dolomite.

[c] Sulfated dolomite after 7 cycles of regeneration.

obvious effect on the rate of $CaSO_4$ reduction.

No deposits of any kind were found, nor was there evidence of ash slagging in any of the runs.

In none of the runs was process heat balance achieved simultaneously with the desired ratio, $(CO + H_2)/(\text{total sulfur}) = 2$. Four of the six runs were strongly exothermic. Table 2 shows calculated process conditions which lead to thermoneutrality and a $(CO + H_2)/(\text{total sulfur})$ ratio of 2. For the process calculations, a carbon burnout of 70 percent (versus 60.3 percent in Run A7) was assumed, to allow the effects of a deeper fluidized bed. The process calculations show that the following $CaSO_4$/coal/air ratios will be required:

$Coal/CaSO_4$ = 72 lb coal/mol $CaSO_4$ fed.

Air/Coal = 27 SCF/lb coal.

Table 2. CALCULATED PROCESS CONDITIONS FOR HEAT BALANCE IN STAGE 1 [a]

	Temp, °F	Lbs	Mols	ΔH, Btu
	Input			
Preoxidized Coal	700	72.45	–	17,600
Sulfated Acceptor				
Inert	1800	56.66	–	27,210
CaO	1800	6.56	0.1173	2,440
$CaSO_4$	1800	136.14	1.000	61,070
Air	100	145.4	5.032	1,400
Heat of Reaction [b]				120,570
Total				230,290
	Output			
MAF Char	1850	15.74	–	10,850
Ash	1850	7.74	–	3,520
Product Acceptor				
Inert	1850	56.66	–	27,990
CaO	1850	18.68	0.3331	7,170
CaS	1850	56.13	0.7781	18,040
$CaSO_4$	1850	0.83	0.0061	390
Product Gas				
H_2O	1850	21.84	1.212	43,440
CO_2	1850	83.71	1.902	40,480
CO	1850	32.38	1.156	15,220
N_2	1850	111.0	3.962	52,900
H_2	1850	0.83	0.412	5,080
S_2	1850	9.68	0.151	2,330
Heat Losses				2,880
Total				230,290

[a] Basis: 1 lb mol $CaSO_4$.

[b] Heat formation at 25°C.
$(CaSO_4)$ in. – $(CaO + CaS + S_2 + CO_2 + CO + H_2O)$ out.

The breakdown of the total oxygen required at process conditions is:

Air	33	percent
Coal	4	percent
$CaSO_4$	63	percent

The air requirement corresponds to 22 percent of stoichiometric air for combustion of the 5 percent preoxidized Ireland Mine coal.

Stage 2 Runs

The feedstock used for these runs was, for experimental convenience, prepared by reducing the $CaSO_4$ to CaS, using CO both as a fuel and reductant at 1750°F. Part of the CO was burned with air to provide the preheat duty for the incoming gas and solids. In the second stage, a portion of the incoming CaS is oxidized via reation (7). The $CaSO_4$ reacts with residual CaS via reaction (8), thereby rejecting sulfur. The overall reaction is highly exothermic and provides sufficient preheat duty for the incoming air and diluent recycled tail gas. During the program, about twice as much diluent (N_2) was used as is required for process heat balance in order to provide a conservative ΔP SO_2 driving force with respect to reaction (8). Run conditions and results are shown in Table 3.

In Run B2 the input air rate was varied to determine the O_2 requirement which gives the optimum sulfur rejection at 1950°F. The optimum conditions appear to be about 95 percent of the theoretical O_2 needed to reject all the sulfur. At higher O_2 inputs, the product acceptor contains $CaSO_4$.

It is not clear whether the lower level of sulfur rejection in run B1 at 1900°F was caused by the lower reaction rates for reaction (8) and/or (7) or by inhibition due to the lower ΔP SO_2 driving force. More data is needed on the kinetics of the second-stage reactions.

Small deposits caused by the $CaSO_4$-CaS transient liquid occurred in all the runs. Their location, appearance, and composition were the same as the deposits which were formed in the CO_2 acceptor process regenerator.[3] In Run B2 the extent of deposit formation was 0.6 percent of the acceptor fed.

After repeated combustion-regeneration cycles, it is likely, as in the CO_2 acceptor process, that the extent of deposit formation will decrease drastically, and that the deposit problem can be tolerated by shutting down periodically to descale the reactor walls.

Sulfur Recovery

Sulfur is recovered by so blending the regeneration off gases from the two stages that the ratio of $(CO + H_2 + H_2S + COS)/(SO_2)$ is equal to or slightly greater than 2. The gas is first passed through a catalytic reductor to effect reaction (5) and the corresponding reaction,

$$2\,H_2 + SO_2 = 1/2\,S_2 + 2\,H_2O. \qquad (9)$$

Small amounts of "trimming" air may be added to the feed gas so that the product gas from the reductor step continues a proper feed to a multistage Claus plant. The equilibriums in reactions (5) and (9) are so favorable that substantially no CO or H_2 remain in the product gas. The Claus feed gas now contains a ratio of $(H_2S + COS)/(SO_2) = 2$.

Figure 3, a schematic flow diagram of the sulfur recovery section, also shows a typical feed gas composition.

A thermodynamic analysis of the sulfur recovery section was made with the basic premises given below.

Table 3. CONDITIONS AND RESULTS FOR SECOND-STAGE RUNS

Run Number	B1	B2(I)	B2(II)	B2(III)
System Pressure, psig	5	5	5	5
Bed Temperature, °F	1900	1950	1950	1950
Acceptor, Mol % of Total Ca				
CaS	73.3	72.3	72.3	72.3
$CaSO_4$	2.4	1.8	1.8	1.8
CaO	24.3	25.9	25.9	25.9
Feed rate, lb/hr	4.64	4.95	4.95	4.95
Inlet Air, SCFH	70.5	64.4	76.4	85.5
Inlet N_2, SCFH	147.0	130.6	127.0	129.0
N_2 Purges, SCFH	3.6	3.6	3.6	3.6
Exit Gas Rate, SCFH	215.0	194.0	202.0	208.0
Exit Gas Composition, Mol %				
N_2	95.76	95.21	94.63	95.52
SO_2	4.24	4.79	5.37	4.48
Outlet Fluidizing Vel., ft/sec	2.42	2.23	2.33	2.39
% Sulfur Rejected	82.1	79.4	93.8	80.1
Mol % Total Ca in Product				
CaO	86.5	84.8	94.8	85.2
CaS	7.3	14.7	4.1	2.0
$CaSO_4$	6.2	0.5	1.1	12.8
Input O_2, % of Theoretical	92.2	79.4	94.4	102.8
Outlet SO_2 Partial Press., atm	0.0560	0.0632	0.0709	0.0592
ΔP Driving Force, atm	0.0680	0.0978	0.0901	0.1018
Bed Density, lb/ft^3	21.2	22.7	22.4	21.7
Solids Retention Time, hr	1.22	1.25	1.21	1.15

Equilibrium is assumed to be established in all stages with respect to the following reactions:

$$CO + 1/2\ S_2 = COS \qquad (10)$$

$$H_2 + 1/2\ S_2 = H_2S \qquad (11)$$

$$2\ H_2 + SO_2 = 1/2\ S_2 + 2\ H_2O \qquad (9)$$

$$2\ CO + SO_2 = 1/2\ S_2 + 2\ CO_2 \qquad (5)$$

$$3\ S_8 = 4\ S_6 \qquad (12)$$

$$S_8 = 4\ S_2 \qquad (13)$$

The equilibrium relationships in the above six reactions, at a given temperature and pressure along with the four elemental balances, completely define the system in each stage.

Experimental data[4] obtained in our laboratories and by others[5] shows that, at least under laboratory conditions, it is very easy to establish equilibrium in all of the above reactions even at very low temperatures, provided an active alumina catalyst is employed.

It is necessary, in order to maintain catalyst activity, to operate above the dew point of the sulfur to prevent its deposition on the catalyst. Each stage was so operated, therefore, that the vapor pressure of sulfur at the outlet temperature was 20 percent greater than its partial pressure in the outlet gas. Sulfur is condensed from the product gas from each stage before feeding the gas to the succeeding stage.

A computer program was set up using an iterative trial-and-error procedure to solve for sulfur recovery and product gas compositions with the above restraints imposed on the system.

The free energy data for reactions (5), (9), (10), and (11) were taken from the Janaf tables.[6] Experimental data[7] was used to define the equilibrium constants for reactions (12) and (13).

The outlet temperatures from each stage, for the illustrative example given, are shown in Figure 3. The total sulfur recovery potential is 98.2 percent. Addition of a fourth stage, operated at 270°F, increases the potential sulfur recovery to 99.3 percent.

Energy and Raw Material Requirements

The relative energy and raw material requirements were evaluated for the two types of fluidized-boiler operations using once-through dolomite and acceptor regeneration with sulfur recovery, respectively. The evaluation is based on the following process conditions:

Sulfur in coal = 4.5 percent
Removal of SO_2 from stack gas = 90 percent
Equilibrium acceptor activity = 0.41, equivalent to 8 percent makeup rate
Second stage = 1950°F
Fluidized boiler = 1800°F
Sulfur recovery section, 1st reactor = 900°F

Table 4. COMPARISON OF RELATIVE ENERGY AND RAW MATERIAL REQUIREMENTS IN FLUIDIZED BOILERS

Requirement	With Regeneration and Sulfur Recovery	Once-Through
Energy as Coal Equivalent[a]		
Coal fed to regeneration	9.16	—
Sensible heat for fresh dolomite	0.39	1.92
Heat to calcine fresh dolomite	0.61	3.01
Less		
Sensible heat regenerated acceptor	0.21	—
Residual char from regeneration burned in boiler	2.22	—
Steam and boiler feed water duty from sulfur recovery	2.20	—
Net	5.53	4.93
Cost - ¢/Ton Coal Burned		
Incremental coal at $6/ton	+33	+30
Fresh dolomite at $2.50/ton	+28	+138
Worth of recovered sulfur at $25/ton	-85	—
Total cost	-24	+168
Relative cost	0	+192

[a] lb/100 lb coal burned in boiler.

Table 4 compares the two systems. The net difference in material costs between the once-through and regeneration processes, about $1.90 per ton coal burned in the boiler, is a powerful incentive to continue the development of the two-stage regeneration process.

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3. Curran, G.P., C.E. Fink, and E. Gorin. U. S. Dept. of Interior, OCR R&D Report No. 16, Interim Report No. 3 on CO_2 Acceptor Process.
4. Consolidation Coal Laboratories. Unpublished Data.
5. Pasternak, R. Brennstoff-Chemie, *50*: 200, 1969.
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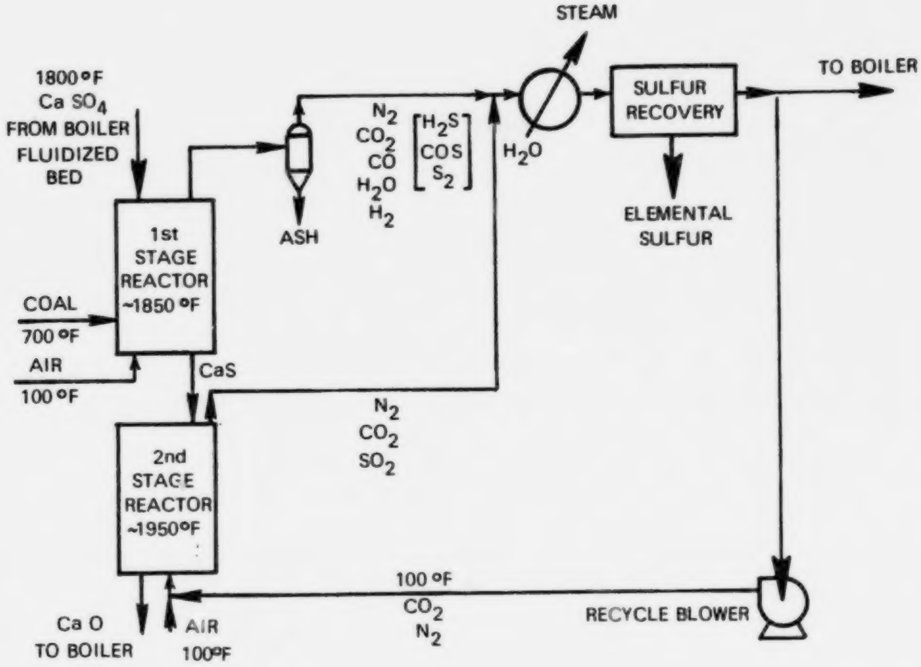

STEAM
TO BOILER
1800 °F
Ca SO_4
FROM BOILER
FLUIDIZED
BED
N_2
CO_2
CO
H_2O
H_2
H_2S
COS
S_2
H_2O
SULFUR
RECOVERY
ELEMENTAL
SULFUR
ASH
1st
STAGE
REACTOR
~1850 °F
COAL
700 °F
AIR
100 °F
CaS
N_2
CO_2
SO_2
2nd
STAGE
REACTOR
~1950°F
100 °F
CO_2
N_2
RECYCLE BLOWER
Ca O
TO BOILER
AIR
100°F

Figure 1. Flow diagram of two-stage regeneration of $CaSO_4$ to CaO.

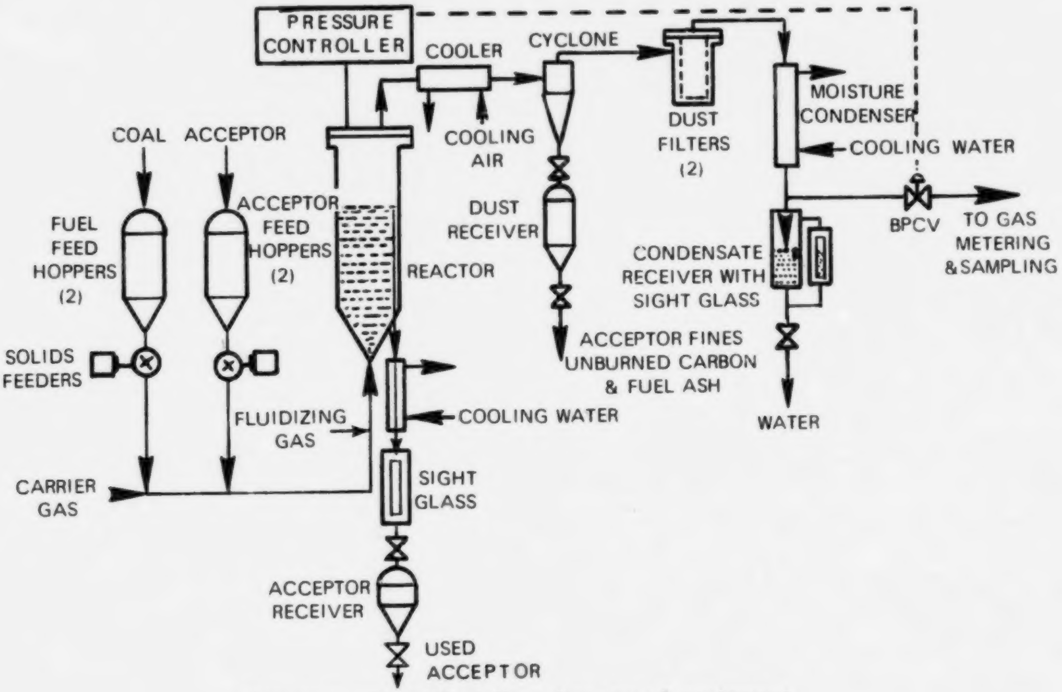
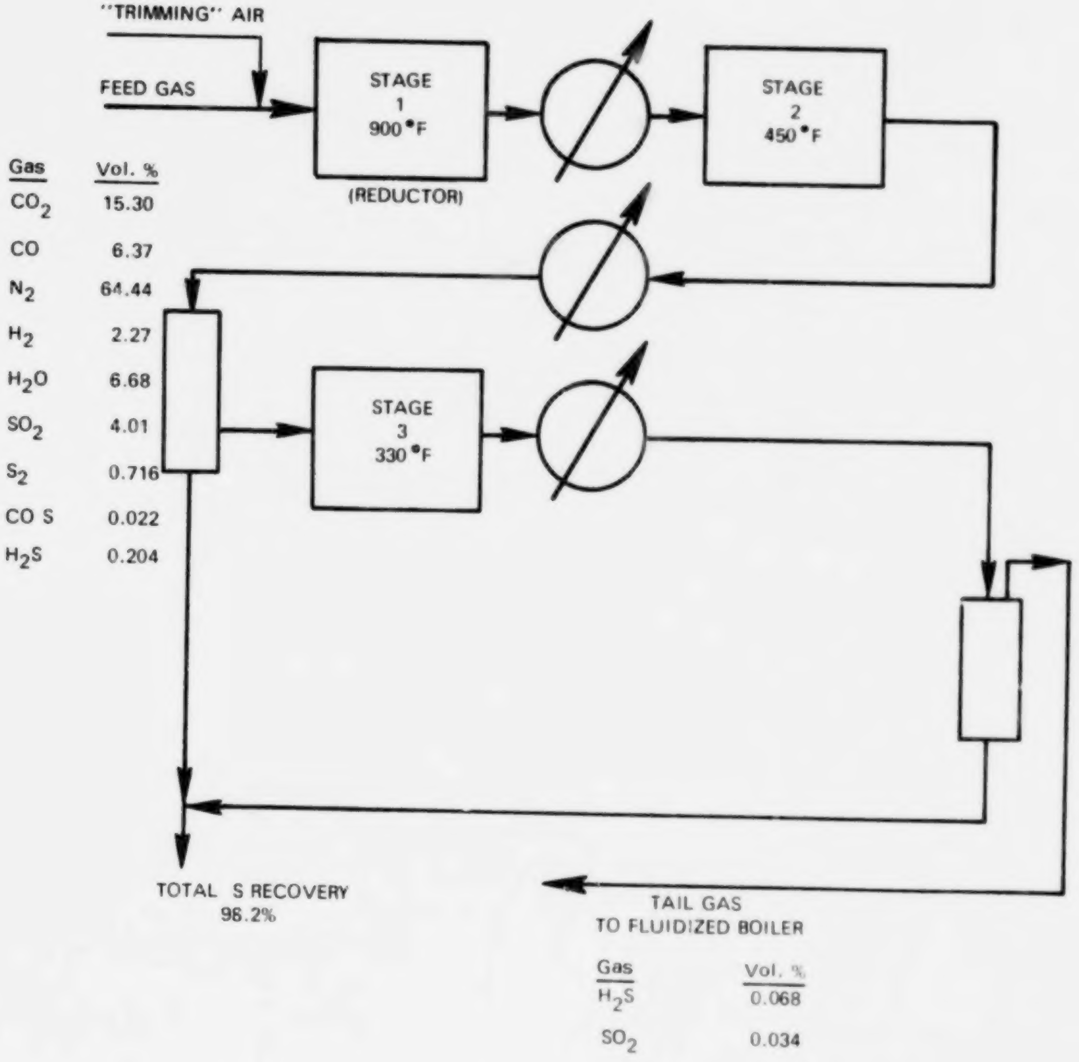

PRESSURE
CONTROLLER
COOLER
CYCLONE
COOLING
AIR
DUST
FILTERS
(2)
MOISTURE
CONDENSER
COOLING WATER
COAL
ACCEPTOR
FUEL
FEED
HOPPERS
(2)
ACCEPTOR
FEED
HOPPERS
(2)
REACTOR
DUST
RECEIVER
CONDENSATE
RECEIVER WITH
SIGHT GLASS
BPCV
TO GAS
METERING
&SAMPLING
SOLIDS
FEEDERS
ACCEPTOR FINES
UNBURNED CARBON
& FUEL ASH
FLUIDIZING
GAS
COOLING WATER
WATER
CARRIER
GAS
SIGHT
GLASS
ACCEPTOR
RECEIVER
USED
ACCEPTOR

Figure 2. Simplified flow diagram of equipment.


"TRIMMING" AIR
FEED GAS
STAGE 1 900°F
(REDUCTOR)
STAGE 2 450°F
STAGE 3 330°F
TOTAL S RECOVERY 96.2%
TAIL GAS TO FLUIDIZED BOILER

Gas	Vol. %
CO_2	15.30
CO	6.37
N_2	64.44
H_2	2.27
H_2O	6.68
SO_2	4.01
S_2	0.716
CO S	0.022
H_2S	0.204

Gas	Vol. %
H_2S	0.068
SO_2	0.034

Figure 3. Sulfur recovery from off gases - - two-stage regeneration of $CaSO_4$.

6. THE FLUIDISED-BED DESULPHURISING GASIFIER

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ABSTRACT

Explanations are offered of the optimum sulphur-absorption temperatures which are observed when fuel oil is combusted in fluidised beds of lime particles, under both oxidising and reducing conditions. In the gasifying case, information is given concerning the effect on desulphurising efficiency of variations in stoichiometric ratio, bed replacement rate, and the mean particle size of the bed material.

Consideration is given to some of the control problems which arise when continuous gasification is attempted; preferred solutions are described.

INTRODUCTION

In a paper presented at the First International Conference on Fluidised-Bed Combustion it was shown that sulphur-containing oils may be desulphurised during combustion within a fluidised bed of lime particles under both oxidising and reducing conditions. In the first case the sulphur is fixed as $CaSO_4$; in the second case it is fixed as CaS. It was reported that in both cases the optimum absorption temperature was in the region of 850°C. It had also been found that regeneration of the reacted bed material was possible in both cases and that high concentrations of SO_2 could be obtained at temperatures in the region of 1050°C, by reducing $CaSO_4$ and by oxidising CaS. Several possible applications of this principle were described and some experimental results were presented relating largely to gasifying conditions.

This paper reviews the progress which has been made in this field since then at the Esso Research Centre at Abingdon.

OPTIMUM TEMPERATURE FOR SULPHUR RETENTION

Thermodynamic considerations indicate that sulphur should be fixed efficiently by lime under both oxidising and reducing conditions at temperatures up to 1100°C. Regeneration is accounted for by the decomposition of calcium sulphite, which is produced (in one case) by the reduction of calcium sulphate, and (in the other case) by the oxidation of calcium sulphide. Figure 1 shows the effect of temperature on the equilibrium partial pressure of SO_2, for the reaction of oxygen with calcium sulphide; in other words, the SO_2 equilibrium concentration with $CaSO_3$. It is quite easy to obtain this curve experimentally and the important point which emerges is that very high SO_2 concentrations may be obtained at temperatures above 1000°C.

Even at 865°C, the equilibrium SO_2 concentration is as high as would be obtained by burning a 4 percent by wt sulphur fuel oil with 5 percent excess air. It follows from this that, under oxidising conditions at temperatures above about 850°C, sulphur cannot be acquired by lime via calcium sulphite but must be fixed as calcium sulphate by direct reaction of calcium oxide with sulphur trioxide.

Figure 2 shows the pronounced optimum temperature which is obtained when sulphur is fixed in a fluidised bed of lime under oxidising conditions as calcium sulphate. It is important to note that this curve relates to lime in the size range 100-200 microns which was 31 percent reacted to calcium sulphate. When a fresh bed was used, the initial absorption efficiency was in the region of 90-95 percent over the whole temperature range of the experiment. It follows from this that we are dealing with a question of kinetics rather than thermodynamics; indications are that the reaction is ash-diffusion limited. Even so some further explanation is required to account for the optimum temperature.

As we have already seen, although there are two possible routes to the formation of calcium sulphate, one of them (via calcium sulphite) may be ruled out on thermodynamic grounds at temperatures much higher than 850°C. This leaves the direct reaction between SO_3 and lime to be considered. Figure 3 shows the equilibrium partial pressure of SO_3 in the combustion products of 4 percent S fuel oil burned with 5 percent excess air, as a function of temperature. Although the formation of SO_3 is favoured by low temperatures, the rate of formation at low temperature can be extremely slow in the absence of a catalyst. It follows from this that the rate of formation of SO_3 and, consequently, its concentration could well reach a maximum level in the region of 850°C. Under ash-diffusion controlled conditions this could result in the optimum SO_2 absorption temperature which has been observed.

Turning now to gasifying conditions, there is in this case a very much simpler explanation of the optimum absorption temperature. It will be appreciated that even under gasifying conditions, the air which enters the bed contains its normal quota of oxygen. Consequently regenerating conditions can occur immediately adjacent to the distributor. As we have seen regeneration is favoured by high temperatures. Therefore it appears that the fall off in the absorption efficiency of the gasifier at high temperatures is due to an increased tendency to regeneration at the distributor, with reabsorption of SO_2 higher in the bed resulting in an internal refluxing of SO_2 within the fluidised bed. Table 1 shows the concentrations of SO_2 in parts per million which may be observed in gas samples drawn from within the bed at various heights above the distributor. The first set of readings relate to a temperature of 850°C; the second, to a temperature of 950°C. It is clear from these results that the sulphur content of the lime in a bed sample can, under some conditions, vary according to the position within the bed from which the sample is taken.

Table 1. CONCENTRATION OF SO_2 AT STATED HEIGHTS ABOVE DISTRIBUTOR[a] AT 30 PERCENT STOICHIOMETRIC AIR

Height, in.	At 850°C, ppm	At 950°C, ppm
1/2	20	1000
4	10	50
8	10	20

[a] Stone sulphur content 6 percent by weight.

INFLUENCES OF OPERATIONAL VARIABLES ON GASIFIER PERFORMANCE

Stoichiometric Ratio

At the first Conference, information was presented concerning the effect of stoichiometric ratio on the desulphurising efficiency of a gasifier. At that time the gasifier was being operated with some degree of pre-combustion; about 25-35 percent of the incoming air was being burned before reaching the distributor. This procedure results in a bed of uniform sulphur content at temperatures in the region of 850°C and enables the sulphur balance to be checked by stone analysis. Since then the apparatus has been operated without underfiring and it has been found that this method of operation is markedly superior.

Figure 4 shows the effect of changing stoichiometric ratios on desulphurising efficiency, with and without underfiring, with 4 percent by wt of sulphur in the 800-micron mean-particle-size bed material. It can be seen that, in the absence of underfiring, the desulphurising efficiency is maintained even at a stoichiometric ratio of 20 percent; whereas, when the bed is underfired, there is a marked drop in desulphurising efficiency at stoichiometric ratios lower than 30 percent. Furthermore the results which were obtained without underfiring were obtained at a superficial gas velocity of 4 ft/sec, as against the 3 ft/sec used with underfiring. This enhanced performance naturally has a considerable impact on the size of the gasifier and therefore on the investment required for a given fuel throughput. The reason for the poor performance in the presence of underfiring is not entirely clear, but the particles get very sooty under these conditions at low stoichiometric ratios whereas, in the absence of underfiring, they remain clean.

Bed Material Replacement Rate

A very important factor bearing on the running costs of the desulphurising gasifier is the rate at which the bed material has to be replaced in order to maintain desulphurising efficiency. In order to obtain information on this subject, we ran several series of cyclic tests each at a different bed replacement rate, both with and without underfiring. The results of just one of these test series are shown in Figure 5. In this case the bed replacement rate was 0.5 wt of lime per unit weight of sulphur; i.e., about a third of the stoichiometric requirement. It can be seen that under these conditions the desulphurising efficiency rapidly bottomed out at about 60 percent. The effect of varying bed replacement rates on desulphurising efficiency can be seen in Figure 6. Although these results were obtained using underfiring, the efficiencies measured without underfiring fall on the same line. It is clear from this curve that virtually 100-percent desulphurising efficiency is obtainable with a bed replacement rate which meets the stoichiometric requirement of the sulphur; i.e., about 1.75 wts of lime per unit weight of sulphur. This result relates to a maximum stone sulphur content of approximately 4 percent by weight.

Particle Size

The effect of particle size on desulphurising efficiency has also been examined. No discernible difference in performance was found when the same stone was compared at mean particle sizes of 810 and 635 microns. Indications are that, due to the highly porous nature of the lime particles, the internal surface contributes most of the reactive surface area; consequently, there is little to be gained by making the mean particle size smaller than the diameter at which bed containment becomes a problem.

An interesting feature of the system is its ability to fix vanadium as well as sulphur. Vanadium may be fixed with virtually 100-percent efficiency under both oxidising and reducing conditions.

DESIGN OF A CONTINUOUSLY OPERATING GASIFIER

With the feasibility of the desulphurising gasifier established, consideration was given to the design problems posed by a practical system; an experimental unit is currently being constructed which will deliver 7 MM Btu of desulphurised fuel per hour on a continuous basis.

Figure 7 is a schematic flow diagram for a continuous unit. On the left-hand side is the gasifying bed in which lime is converted into calcium sulphide; on the right-hand side is the regenerator in which calcium sulphide is oxidised by air to lime and SO_2: bed material is circulated between the two units. If the gasifier is to function efficiently, it is necessary to maintain the temperature of the gasifying bed in the region of 850°C and the temperature of the regenerating bed in the region of 1050°C. Should the temperature of the regenerating bed fall below 800°C, recarbonation will occur, lowering the temperature of the regenerator. If the gasifying temperature rises above 900°C, the desulphurising efficiency will be reduced. In the case of the regenerator, too low a temperature will result in the formation of calcium sulphate and a buildup of sulphur in the bed material; too high a temperature may tend to deactivate the lime.

A method of controlling the temperatures of the two beds has been chosen which involves no heat losses and no heat-transfer surfaces. In the case of the gasifying bed, it is proposed to control the temperature by recycling some of the flue gas formed by complete combustion back to the inlet of the air blower. This necessitates a separate air supply for the regenerator. In the case of the regenerator, the temperature will be controlled by varying the circulation rate of the bed material. The rate of heat release in the regenerator is determined by the rate at which air is supplied: it will normally match the rate at which sulphur is acquired by the gasifier. The effect of increasing the flow rate of bed material will be to reduce its sulphur content and, hence, the amount of heat generated within the regenerator per unit weight of lime. Since the lime enters the regenerator at a temperature of 850°C and is heated to a temperature of 1050°C, there must clearly be a specific bed transfer rate for any particular set of operating conditions at which the regenerator temperature is in equilibrium at the desired level. It follows from this that an accurate and reliable method for controlling bed transfer rates is an essential requirement for successful operation.

The method which has been chosen is gravitational dense phase transfer. Each bed container is provided with a catchment pocket slightly above the surface of the bed; each of these pockets communicates via an almost vertical duct with the side wall of its neighbor at a point slightly above the level of the distributor. There is a short horizontal section at the bottom of each duct so that, in the absence of any external stimulus, the two transfer ducts simply fill with static bed material. An arrangement of this type is shown in Figure 8 which is a schematic layout of the first bed transfer test rig. Here you can see the two transfer ducts and their horizontal delivering sections. Each horizontal connection has a suitable perforated tube running parallel to its axis. When gas is introduced via one of these tubes, the particles in the horizontal duct are fluidised and expelled; they are replaced by gravitational flow down the vertical duct. As soon as the gas ceases to flow, so do the solids. By pulsing the gas flow and varying the frequency of the pulses, it is possible to closely control the rate at which bed material is transferred. Very little gas is required to operate the system. In the original test rig it was possible to shift about 2 pounds of bed material per cubic foot of gas. In view of the possible variation of sulphur content throughout the depth of the bed, the transfer of material from the top of each bed to the bottom of the other is advantageous from an operational point of view. The dense packing in each duct, together with the continuous gas bleed which is required to prevent blockage of the perforations in the activating tubes, ensures that the leakage of gas between compartments is minimal.

The experimental gasifier has been designed as a monolithic structure in castable refractory which is insulated, from the walls of the mild steel casing which contains it, by a layer of vermiculite fondu. The overall dimensions of the unit are about 4 feet square by 8 feet high. In order to check the operation of the bed transfer system and the cyclones, the full-scale cold rig (shown in Figure 9) has been built in mild steel and perspex. The large tank represents the gasifying compartment: the central cylinder flanked by the two cyclones represents the regenerator. The perspex ducts allow observations to be made of the flow of bed material which can of course be measured by weight. The bed material which is being used in these tests is crushed brick with the same size distribution and bulk density as the lime. The results which have been obtained have been used as a basis for the experimental gasifier which is now being constructed as part of a contract with NAPCA for development of the chemically active fluid-bed gasifier.

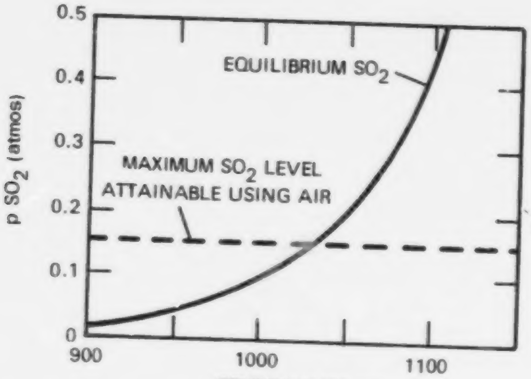

Figure 1. Effect of temperature on the equilibrium SO_2 partial pressures for the reaction of oxygen with calcium sulphide.

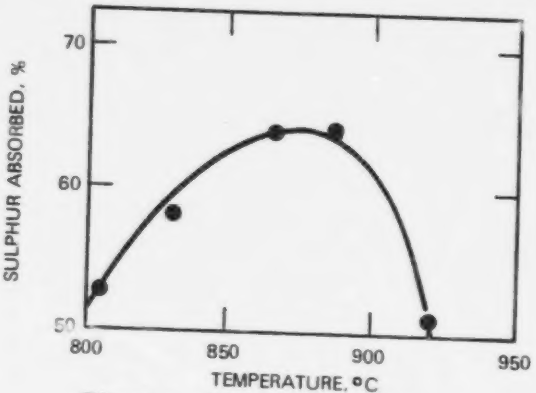

Figure 2. Optimum temperature for sulphur oxide absorption by lime (31% of bed reacted).

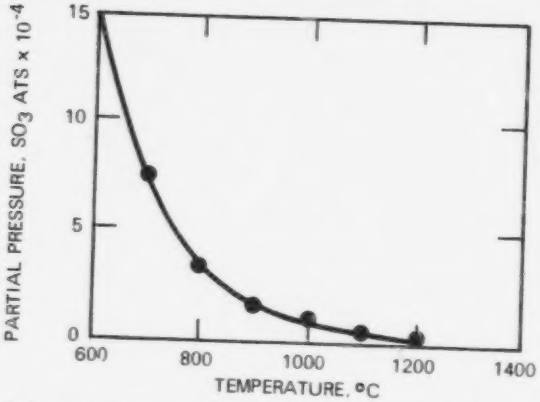

Figure 3. Equilibrium partial pressure of SO_3 in combustion products of 4% S fuel oil burned with 5% excess air.

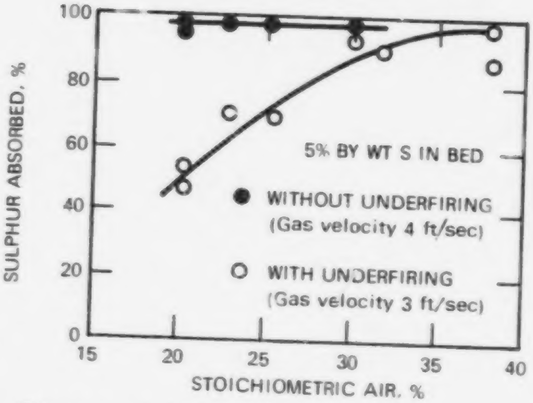

Figure 4. Stoichiometric air rate with and without underfiring.

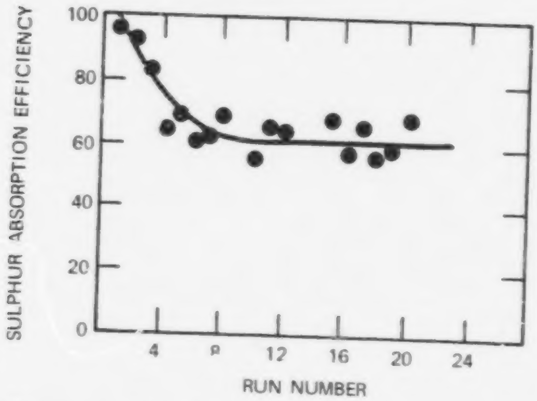

Figure 5. Bed ageing effect replacement rate 0.5 wt CaO/wt S.

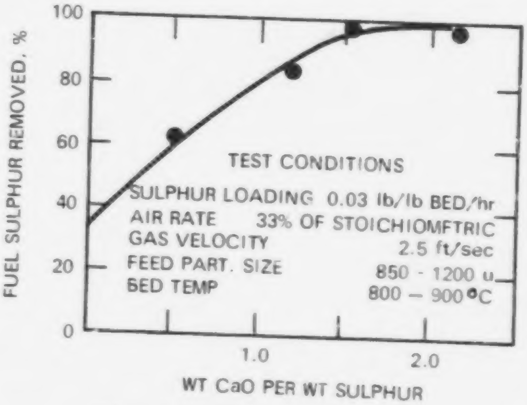

Figure 6. Effect of makeup rate on desulphurising efficiency.

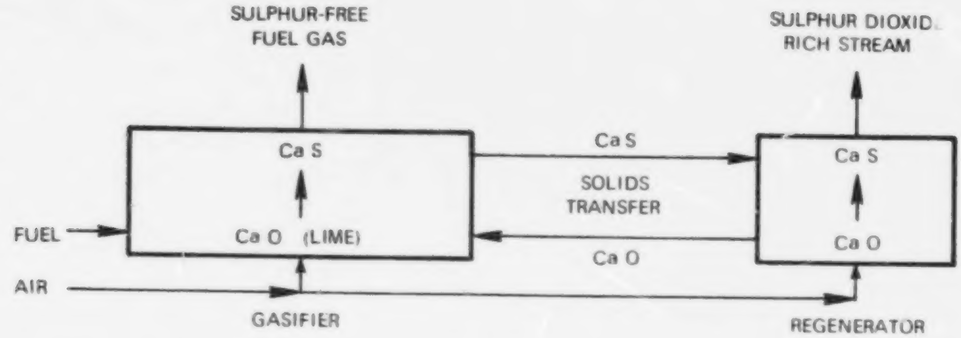
SULPHUR-FREE FUEL GAS
SULPHUR DIOXID RICH STREAM
Ca S
Ca S
SOLIDS TRANSFER
Ca S
FUEL
Ca O (LIME)
Ca O
Ca O
AIR
GASIFIER
REGENERATOR

Figure 7. Chemically active fluid-bed gasifier.

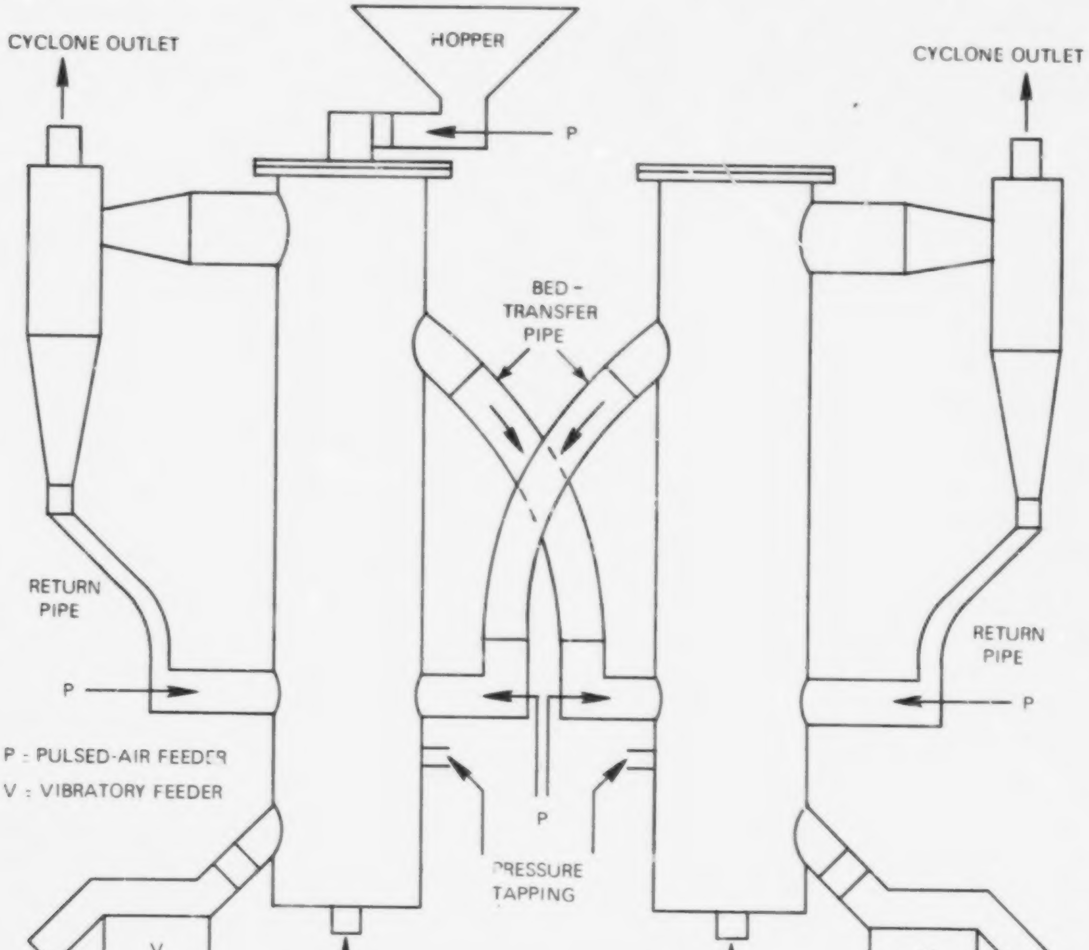
CYCLONE OUTLET
HOPPER
P
CYCLONE OUTLET
BED - TRANSFER PIPE
RETURN PIPE
RETURN PIPE
P
P
P : PULSED-AIR FEEDER
V : VIBRATORY FEEDER
P
PRESSURE TAPPING
V
V
AIR SUPPLY

Figure 8. Test rig for bed-transfer trials.

SESSION III:

Gasification to Desulfurize Coal

SESSION CHAIRMAN:

Mr. J. W. Eckerd, USBM, Morgantown

1. PRODUCTION OF LOW-SULFUR BOILER FUEL BY TWO-STAGE COMBUSTION — APPLICATION OF CO_2 ACCEPTOR PROCESS

G. P. CURRAN, C. E. FINK, AND
E. GORIN

Consolidation Coal Company

ABSTRACT

A modification of the CO_2 acceptor process is presented as a method for the production of low-sulfur boiler fuels; i.e., low-sulfur char and/or low-sulfur producer gas. Particular emphasis is given to total gasification of Eastern coals for the production of low-sulfur producer gas, which could be utilized for the production of clean power in new combined-cycle plants. Experimental data is given on acceptor life under projected operating conditions

INTRODUCTION

Considerable interest is developing in the production of clean power from coal by means of a combined-cycle power plant. The first step necessary in such a plant is the generation of high-pressure producer gas by the gasification of coal. Recently[1] construction of a 170,000 KW plant of this type in Germany was announced by Steinkohlen-Electrizität AG. The plant will adapt the well known Lurgi pressure gasification process to production of high-pressure producer gas. Surprisingly, although no process is provided for desulfurization of the gas, the incentive for construction of the plant is stated[1] to be its lower investment cost in comparison with that of a conventional power plant. If the economics can be confirmed, the combined-cycle plant opens up the prospect of producing clean power from coal at a possible lower cost in new plants than in conventional plants equipped with stack gas scrubbing equipment.

The most critical factor in the development of such a new power cycle is the availability of a satisfactory process for the production of low-sulfur, high-pressure producer gas. The Lurgi process, which must include facilities for desulfurization of the gas, is the only such process commercially available today. Eastern bituminous coals, however, for the most part, offer difficulties as feedstocks to the Lurgi process because of coking problems. Furthermore, it would be desirable to purify and desulfurize the gas while it is hot, as part of the gasification process, in order to improve thermal efficiency of the cycle. High thermal efficiency in the combined-cycle plant can only be obtained if the gas can be cleaned and desulfurized hot.

The CO_2 acceptor process is now undergoing intensive development[2] as a means of producing high-Btu pipeline gas from coal. It is the purpose of this paper to show that many of the features of this process and the experience gained in its development can be utilized to speed the development of an analogous process for the production of clean, high-pressure producer gas.

A contract, with the same title as this paper, has been signed with NAPCA to accelerate research on this application of the CO_2 acceptor process. The work reported here concerns both some precontract work carried out by Consolidation Coal Research and some initial results of work carried out under the contract.

DESCRIPTION OF THE PROCESS

The proposed process is in reality a two-stage, air-blown, fluidized gas producer. The simple air-blown, pressurized, fluidized gas producer itself is not a developed process; many problems have been associated with its development. These potentially can be overcome by use of the present process. The advantages of the present process are:

1. In situ desulfurization of the generated gases.
2. Improved efficiency of carbon utilization.
3. Improved operability by control of the ash-fusion problem.

The first advantage stems from the well known sulfur-acceptor properties of the lime acceptor. The second and third advantages arise because of the two-stage nature of the process. The final combustion of carbon is conducted, separate from the gasification reaction, in the presence of the endothermic lime calcining reaction.

Figure 1 shows one version of the process presently being analyzed as part of the feasibility study under the NAPCA contract. The coal is preoxidized in a separate air-blown fluidized vessel operated at 750-800°F.

A computer program, consistent with thermodynamic restraints imposed on the process, gives the heat and material balance of the system illustrated in Figure 1. The program is broad enough to encompass variations in the process; e.g., producing low-sulfur char as a co-product with the low-sulfur producer gas. The char-to-gas ratio can be varied over wide limits.

Table 1 is a condensed version of the heat and material balance for one-spot condition. This corresponds to the situation where complete conversion to producer gas is effected; i.e., no char byproduct occurs. The heat and material balance relationships given for the gasifier correspond to gasification of 65 percent of the fixed carbon in the feed coal.

The process consists of two main process steps–gasification and acceptor regeneration–connected in series. The gasifier is operated at 15-20 atmospheres (atm) pressure at 1700-1775°F. Preoxidized coal is fed to the gasifier along with excess air. The char bed is fluidized by steam and recycle gas from the regenerator operation after it has been processed for sulfur recovery in the modified Claus plant that is described later. Calcined dolomite from the regenerator is fed to the top of the gasifier.

The endothermic heat of gasification in the gasifier is supplied, both by the partial combustion of carbon with air, and by the exothermic heat of absorption of the CO_2 by the calcined dolomite.

It is necessary, in order to supply chemical reaction heat to the gasifier, that a driving force for absorption of CO_2 by the lime be present. Because the acceptor falls through the fluidized char in a "showering" action, it is only necessary that the driving force be present in the bottom part of the bed. At the gasifier temperature chosen in the Table 1 illustration, 1700°F, such a driving force for CO_2 does exist at the bottom of the gasifier: $P_{CO_2} = 2.9$ atm versus the equilibrium value of 1.6 atm.

Another essential feature of the gasifier operation is simultaneous desulfurization of the producer gas in situ, via the acceptor reaction:

$$CaO + H_2S = CaS + H_2O \qquad (1)$$

$$K = P_{H_2O}/P_{H_2S} = 525 \text{ at } 1700°F$$

Table 1. SUMMARIZED HEAT AND MATERIAL BALANCE[a]

Material	Preoxidizer and producer		Material	Regenerator	
	°F	mol		°F	mol
Input					
Coal	60	--	Char	1700	--
C		5.812	C		1.223
H (as H_2)		2.381	H		0.054
O		0.475	Ash (lb)		12.30
S		0.1341			
Ash, lb		12.30			
Steam	1200	3.745			
Coal moisture	60	0.354			
Dry air	398	7.174	Dry air	398	5.422
Moisture	398	0.031	Moisture	398	0.023
Claus plant tail gas	1200		Lift gas	280	
CO		0.105	CO		0.011
CO_2		1.958	CO_2		0.124
N_2		4.284	N_2		0.281
H_2O		0.075	H_2O		0.002
H_2		0.002	Acceptor	1700	
Acceptor	1885		MgO·CaO		6.0318
MgO·CaO		7.0861	MgO·CaO_3		0.8904
			MgO·CaS		0.1285
			$MgCO_3$·$CaCO_3$ (make-up)		0.0354
Output					
Char	1700		Char	1885	
C		1.223	C		0.122
H		0.054	Ash (lb)		12.30
Ash, lb		12.30	Gas	1885	
Gas	1700		CO		0.172
CH_4		0.277	CO_2		2.024
H_2		3.288	N_2		4.565
CO		3.523	H_2O		0.075
CO_2		1.956	H_2		0.004
H_2O		2.617	SO_2		0.0300
N_2		9.951	S_2		0.0487
NH_3		0.047	H_2S		0.0005
H_2S		0.0050	COS		0.0006
Acceptor[b]	1700		Acceptor	1885	
MgO·CaO		(0.0303) 6.0318	MgO·CaO		7.0861
MgO·$CaCO_3$		(0.0045) 0.8904			
MgO·CaS		(0.0006) 0.1285			

[a] Basis: 100 lb dry coal
15 atm system pressure
14 atm clean gas delivery pressure

[b] Numbers in parentheses are for acceptor discarded in order to maintain acceptor activity.

$$\text{Cold gas efficiency} = \frac{\text{Gross heating value of cold gas}}{\text{Gross heating value of coal feed}} = 74.7\%.$$

The gas composition given in Table 1 presumes that equilibrium is established in reaction (1). The potential extent of desulfurization under the operating conditions given in Table 1 is thus 96 percent; i.e., the sulfur in the producer gas product is only 4 percent of that in the feed coal.

Acceptor regeneration is the second principal process step. The acceptor is regenerated by burning fuel char in an air-fluidized bed. The fuel char is the gasification residue and its combustion provides the heat to carry out the endothermic calcination reaction:

$$CaCO_3 = CaO + CO_2 \qquad (2)$$

The regenerator thus serves as a receptacle for gasification residue that can be efficiently used as fuel. This procedure significantly improves the total carbon gasification efficiency over that of an air-blown fluidized producer.

Another feature of the regenerator operation is that combustion of char is conducted concurrently with the endothermic calcination reaction (2). This feature minimizes the difficulties due to ash fusion that are usually encountered in combustion of char in fluidized beds and that are particularly severe when the operation is conducted under pressure.

Another important process that occurs in the regenerator is rejection of sulfur via the overall roasting reaction:

$$CaS + 3/2\ O_2 = CaO + SO_2 \qquad (3)$$

Reaction (3) is in reality the composite of two separate reactions:

$$CaS + 2\ O_2 = CaSO_4 \qquad (4)$$

$$CaS + 3\ CaSO_4 = 4\ CaO + 4\ SO_2 \qquad (5)$$

The chemistry of the sulfur cycle in the regenerator[2c] has been extensively studied in work done for the Office of Coal Research in development of the CO_2 acceptor process.

The material balance of Table 1 provides for a driving force sufficient to eliminate SO_2 via reaction (5); i.e., $P_{SO_2} = 0.065$ atm versus the equilibrium value of $P_{SO_2} = 0.090$ atm at the regenerator operating temperature of 1885°F. This driving force is sustained by operating at slightly reduced conditions in order to generate enough CO to reduce SO_2 by the reaction:

$$2\ CO + SO_2 = 1/2\ S_2 + 2\ CO_2 \qquad (6)$$

Note that more than half the sulfur issuing from the regenerator is in the form of elemental sulfur.

There appears to be a theoretical difficulty in this method of operation; i.e., at the CO levels used, calcium sulfate would not seem to be present to effect reaction (5) because it would be reduced by the reaction:

$$CaSO_4 + 4\ CO = CaS + 4\ CO_2 \qquad (7)$$

In practice, however, an environment of oxidation sufficient to form $CaSO_4$ is present throughout most of the regenerator bed, and reaction (7) occurs too slowly to reduce all of the calcium sulfate at the bed outlet, perhaps because of a slow rate of diffusion of CO into the acceptor pores. Verification of this method of operation is described later in this paper.

Sufficient CO is also present in the regenerator off gases to permit reduction of the SO_2 to elemental sulfur by a modified multistage Claus process. The process is essentially the same as that described in another paper[4] presented to this conference. Table 1 gives the composition of the Claus plant tail gas.

Important considerations in the development of a process of this type are acceptor life and cost. Some information has been developed experimentally under conditions similar to those proposed for this process; however, more is required. The data available to date will be presented later in the paper. Table 2 gives the estimated acceptor makeup rate and cost under the conditions proposed for the process. The makeup cost is approximately 1.3¢/MM Btu of product gas.

Table 2. DOLOMITE MAKEUP COST AND SULFUR REJECTION

Makeup rate, mol $MgCO_3 \cdot CaCO_3$/mol $MgO \cdot CaO$ circulated, %	0.5
Lb raw dolomite/100 lb dry-coal feed	7.41
Makeup cost, ¢/10^6 Btu[a] at $3.25/ton	1.27
Sulfur distribution	
Product gas, %	3.7
Acceptor, %	96.3
Spent acceptor composition	
CaO, mol, %	85.55
$CaCO_3$	12.63
CaS	1.82

[a] Gross heat of combustion of product gas.

The literature does not provide suitable kinetic data to properly size and define the optimum operating conditions in the gasifier. Some limited integral rate data is presented later in this paper; however, more is required to further develop the process.

EXPERIMENTAL PROCEDURE

The equipment and operating procedure have both been fully described in the report to the Office of Coal Research.[2c] Briefly, the gasifier was a 70-in. long, thin-walled vessel, with a 4-in. ID. The fluidized bed height was controlled at 40-in. by an overflow weir. The vessel was contained, along with the electrical heaters and insulation, inside a pressure shell. CO_2 was used as pressure-balancing gas. The 38-in. long regenerator was of similar construction, with a 2-in. ID; the fluidized-bed height was controlled at 22 in. by the overflow weir. These two reactors were designed to operate at temperatures up to 1950°F and pressures up to 300 psig. All vessels and internals were Type 310 stainless steel except the regenerator thermowell, which was Hastelloy X. The acceptor, sized to 16 x 28 mesh, was dolomite from the Tymochtee formation in Western Ohio.

Calibrated rotary feeders controlled the flow rates of three of the solids: (1) gasifier feedstock (char or coal), (2) regenerator fuel char, and (3) acceptor feed to regenerator. These solids were fed from dual lockhoppers through pneumatic transfer lines. The acceptor circulated continuously between the two vessels. Acceptor flowed from the regenerator to the gasifier by gravity, through a water-cooled standleg. The acceptor particles showered down through the fluidized bed of char and segregated as a separate fluidized bed in the 2-1/2 in. ID "boot" of the gasifier. The acceptor was then withdrawn through a water-cooled standleg and a rotary device similar to the solids feeders, and was discharged periodically from dual lockhoppers situated below the rotary device.

A fixed investory of acceptor, corresponding to 18 pounds of raw dolomite, was used in each run. No fresh makeup was added. The acceptor discharged from the bottom of the gasifier was sampled periodically; its activity was determined by a special assay. Thus, the relationship between activity and the number of calcining-recarbonation cycles undergone by the acceptor could be determined.

In the regenerator, the heat required to calcine the acceptor was supplied by the combustion of char, which was partially gasified material from a previous run. The fuel-char ash was elutriated from the fluidized bed of acceptor and was removed by an external cyclone.

The inlet gas to both vessels contained recycled product gas, which was used to simulate the partial pressures that would exist near the top of the tall fluidized beds in the commercial process. Hydrogen and CO_2 were added to the gasifier recycle gas to adjust the exit partial pressures to the desired values. This inlet dry gas was then passed through a saturator in which the water temperature was controlled to give the desired inlet-steam partial pressure. The regenerator inlet gas was primarily recycled gas and air. CO_2 or N_2 was added to adjust the outlet CO_2 partial pressure to the desired value. The product gas from both vessels was analyzed frequently by gas chromatography.

RESULTS AND DISCUSSION

The experiments reported here were carried out under conditions similar to those for the gasifier of the process under discussion, although they were originally designed for investigation of the process as a "CO maker." The acceptor life and integral rate data generated are therefore similar, but not identical, to what might be expected in practice with the process described above. In general, conditions (from the point of view of acceptor life) were more severe than those projected for the process under discussion; i.e., higher temperatures were used in the gasifier and regenerator, and lower steam pressures were used in the gasifier.

Tables 3 and 4 show the conditions and results from the runs made during the program. Table 3 shows the conditions and results of Run K1, which was made: (1) to demonstrate the operability of the process, (2) to produce fuel char, and (3) to obtain some practical gasification kinetics data. The conditions and results for a typical acceptor life study run are shown in Tables 3 and 4 for the gasifier and regenerator, respectively.

Table 3. GASIFIER RUN CONDITIONS AND RESULTS

	Run No.	
	K1	L2
Cycle	--	15
System pressure, atm	18	18
Acceptor	--	Tymochtee-6 Dolomite
Size, mesh	--	16 x 28
Activity	--	0.39
Circulation rate, lb/hr (raw basis)	--	8.18
Gasifier		
Temperature, °F	1770	1775
Feedstock	Colstrip II Subbituminous Dried at 400°F	Disco char Carbonized Dried at 950°F
Feed rate, lb/hr (MF basis)	5.74	2.95
Product char rate, lb/hr	3.20	1.86
Recycle fluidizing gas, scfh	161.4	232.3
Inlet steam, scfh	2.5	26.5
H_2, scfh	0	0
CO_2, scfh	51.2	54.3
N_2, scfh	0.5	7.2
Total inlet gas, scfh	215.6	320.3
Inlet fluidizing velocity, ft/sec		
Acceptor layer	--	0.540
Bottom of char bed	0.165	0.246

Table 3 (continued) GASIFIER RUN CONDITIONS AND RESULTS

	Run No.	
	K1	L2
Inlet gas partial pressures, atm		
H_2O	0.21	1.49
H_2	2.09	1.24
CH_4	0.56	0.08
CO	7.86	6.01
CO_2	6.57	6.24
N_2	0.68	2.90
Others	0.03	0.08
Product gas, scfh	116.6	103.7
N_2 purges, scfh	4.9	12.7
Condensate, scfh	20.4	27.3
Total outlet gas, scfh	293.4	352.5
Outlet fluidizing velocity, ft/sec	0.225	0.271
Outlet gas partial pressures, atm		
H_2O	1.25	1.39
H_2	2.63	1.63
CH_4	0.70	0.11
CO	9.86	7.89
CO_2	2.95	4.23
N_2	0.50	2.64
Others	0.11	0.11
Product gas composition, mol %		
H_2	15.54	9.51
CH_4	4.14	0.65
CO	58.37	45.98
CO_2	17.01	24.37
N_2	4.75	19.15
Others	0.19	0.34
Product char particle density, lb/ft^3	52.7	71.8
Char bed density, lb/ft^3	25.0	27.4
Char bed weight, lb	6.21	6.87
Retention times		
Solids, hr	1.94	3.69
Vapor, sec	15.1	11.4
Gasification rate, $\frac{\text{lb carbon gasified}}{(\text{lb C in bed} \times \text{min})} \times 10^4$	43.8	25.7
Carbon burnout, %	32.2	36.2
Equilibrium CO_2 partial pressure, atm	2.53	2.62
CO_2 partial pressure driving force, atm	--	1.61
Char product		
H, wt %	0.35	0.30
C	83.23	76.17
N	0.41	0.64
O	0	0
S	1.00	0.29
Ash (diff.)	15.01	22.60

Table 4. REGENERATOR RUN CONDITIONS AND RESULTS

	Run No. L2
Cycle	15
System pressure, atm	18
Acceptor	Tymochtee dolomite
Size, mesh	16 x 28
Circulation rate, lb/hr (raw basis)	8.18
Activity	0.39
Regenerator	
Temperature, °F	1940
Fuel char	Colstrip Subbituminous char
Size, mesh	28 x 150
Feed rate, lb/hr	1.58
Overhead solids rate, lb/hr	0.82
Recycle gas, scfh	182
Inlet gas	
N_2, scfh	45
CO_2, scfh	28
Air, scfh	100
Total inlet gas, scfh	355
Inlet fluidizing velocity, ft/sec	1.15
Inlet gas partial pressure, atm	
O_2	1.06
CO	0.44
CO_2	3.73
N_2	12.77
Outlet gas	
Product gas, scfh	197
N_2 purges, scfh	15
Total outlet gas, scfh	364
Outlet gas partial pressures, atm	
O_2	0
CO	0.89
CO_2	4.69
N_2	12.42
Recycle gas composition, mol %	
O_2	0
CO	4.75
CO_2	25.01
N_2	70.24

Table 4 (continued). REGENERATOR RUN CONDITIONS AND RESULTS

	Run No. L2
Equilibrium CO_2 partial pressure, atm	7.60
CO_2 partial pressure driving force, atm	
Inlet	3.87
Outlet	2.91
Run duration, hours	26.5
Run duration, cycles	25.3
Lb/fuel char fed during run	41.9
Carbon burnout, %	63.2
Overhead material	
H, wt %	0.29
C	54.97
N	--
O	--
S	3.39
Ash (diff.)	41.35
Input char	
H, wt %	0.62
C	77.55
N	0.37
O	0.0
S	0.64
Ash (diff.)	21.44

Operability

No serious operability problems were encountered in using the CO_2 acceptor process to make CO, using sub-bituminous coal as gasifier feed-stock. In Run K1, raw Colstrip sub-bituminous coal that had been dried at 400°F was fed to the gasifier at 1770°F: no coking occurred. The char product particles were not rounded or swollen. In contrast, slight preoxidation (4 percent) of the sub-bituminous coal was required to avoid coking in the hydrodevolatilizer of the conventional CO_2 acceptor process, where H_2 partial pressure was much higher and CO_2 partial pressure was much lower.

However, the process was not operable when preoxidized Pittsburgh seam bituminous coal was used as feedstock.

Two attempts were made to feed Ireland coal that had been preoxidized to levels of 5.5 and 13 percent, respectively, into a bed of Disco char: no massive coking occurred with either feedstock, but the coal became plastic as it entered the bed, and cemented together the char particles. The agglomerates that were formed, although tenuous, plugged the boot and forced a shutdown.

In the actual process as proposed (Figure 1), coking is prevented by feeding the preoxidized coal into the gasifier entrained in a stream of gas containing free oxygen, allowing additional pretreatment to take place at the point of introduction into the gasifier. The efficacy of this method was demonstrated on a 30 ton-per-day pilot-scale unit at the Consolidation Coal Co. Research Center in 1950.

During Run K1, where the vapor retention time was 15 seconds, there was no overt tarry material in the gasifier product gas; however, there was some residual naphthalene. Most of the naphthalene condensed as fog and was collected in glass-wool filters installed downstream of the primary and secondary product gas coolers. Some of the naphthalene condensed in the primary cooler.

No deposits of any kind appeared in the gasifier during Run K1. In the acceptor life study runs where Disco char was used as feedstock, however, slight gasifier deposits occurred, as they had in the earlier work at 1600°F.

Gasification Kinetics

The gasifier was operated with recycled product gas to simulate an upper section of a tall fluidized bed. The feedstocks were either raw coal or partially devolatilized Disco char (produced from Pittsburgh seam coal). Thus, the measured masification rates, shown in Table 2, include carbon-bearing gases formed during pyrolysis and devolatilization. Comparison of the carbon burnout for Run K1 with that for an earlier run, in which Colstrip coal was fed to the conventional CO_2 gasification process,[2c] shows that at the CO_2 and CO partial pressures used, essentially no gasification of fixed carbon occurred in Run K1 and that the yield and product distribution represent only the pyrolysis and devolatilization reactions at the modified process conditions.

The relatively poor kinetics in Run K1 is apparently due to very strong inhibition by CO at the high CO/CO_2 ratios employed in this run. The equilibrium constant in the "Boudouard" reaction is:

$$\frac{(P_{CO})^2}{P_{CO_2}} = K = 100$$

at the conditions used. The K_{exp} at the outlet conditions of Run K1 is about 33. It is clear, therefore, that total inhibition by close approach to equilibrium is not in itself the reason for the poor kinetics.

The integral rates observed with Disco char are representative of what may be achieved with bituminous coal or char feedstocks. They are marginally acceptable for the process under discussion. Because the steam partial pressure would be higher in the producer-gas operation, higher gasification rates are anticipated.

Operation of the Regenerator

The regenerator was operated at a nominal level of 4.5 percent CO in the outlet gas to ensure that no difficulty would be caused by the $CaS\text{-}CaSO_4$ transient liquid. To achieve this conservative CO level with fuel chars having a wide range of particle diameter—28 x 150 mesh versus 48 x 65 mesh used in earlier work —a higher carbon investory was required in the regenerator bed. Therefore, the carbon burnout was lower than that at comparable bed expansions in the earlier work.

Char Desulfurization and Sulfur Cycle

In Run K1 the total sulfur rejected from the Colstrip coal in the gasifier was 35 percent. This low level is typical of the conventional CO_2 acceptor process using low-ranking Western coals. The high lime content of the ash in these coals causes "self-acceptance," with the result that most of the sulfur is carried to the regenerator in the form of CaS. In the life study runs with Disco char, which has low lime content in the ash, sulfur rejection was at the high level of 83-84 percent. The sulfur content of the product char was only 0.29 wt. percent versus 1.16 wt. percent in the feed. The above desulfurization was effected with a hydrogen partial pressure of

1.6 atm. The results are roughly in accord with prior work[3] on desulfurization of low-temperature chars from bituminous coals.

The sulfur content of the gasifier off gases was not determined. However, if equilibrium were approached in the acceptor reaction,

$$CaO + H_2S = CaS + H_2O,$$

the H_2S content of the gas would have been about 0.02 mol percent.

In the life study runs, the rejected sulfur was equivalent to the conversion to CaS of 1.8 mol percent of the CaO content of the acceptor on each pass through the gasifier, a level considerably higher than in any run during the conventional CO_2 acceptor process studies. Analyses of the acceptor, sampled periodically from the gasifier, showed that all of the sulfur had been rejected in the regenerator, even at the relatively high level (4.5 percent) of CO in the exit gas.

Acceptor Activity

Figure 2 shows acceptor activity, measured preiodically during the runs, as a function of the number of calcining-recarbonation cycles. The activity is defined as the mol percent of CaO that is active as a CO_2 acceptor in the recirculating inventory. Equilibrium activities ($\bar{R}$) calculated from the data in Figure 2, are shown below as a function of fresh acceptor makeup rate. The method for calculating the equilibrium activity has been given previously.[2c]

Makeup, %	$\bar{R}$
1	0.143
2	0.244
3	0.320

Note that the figure used for acceptor makeup in Table 2 was only half of the figures above, derived from the experimental data presented here. The lower value is justified by the fact that the process is projected to operate under milder conditions than those under which the above data was obtained. Correlations developed in prior work[2c] on development of the CO_2 acceptor process were used to justify the lower makeup cost.

The relative influences of gasifier and regenerator bed temperatures, of steam and H_2 partial pressures, and of the extent of conversion of CaS on acceptor activity remain to be determined over wider ranges of these variables than were used in the brief study reported here.

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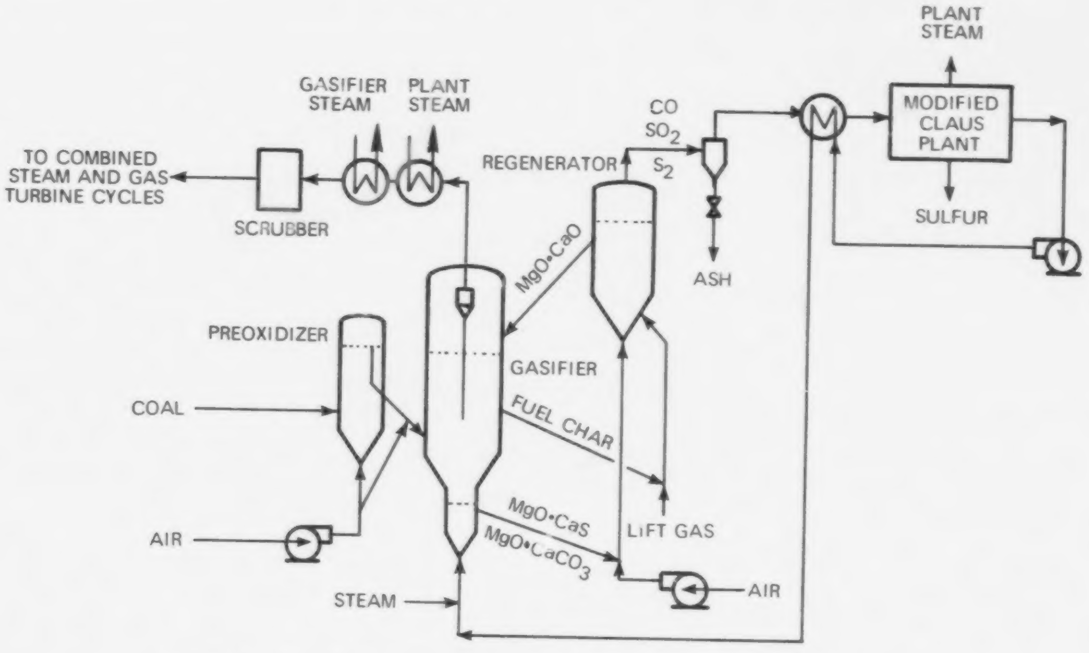
PLANT STEAM
GASIFIER STEAM
PLANT STEAM
CO
SO_2
S_2
TO COMBINED STEAM AND GAS TURBINE CYCLES
SCRUBBER
REGENERATOR
MODIFIED CLAUS PLANT
SULFUR
ASH
MgO•CaO
PREOXIDIZER
GASIFIER
COAL
FUEL CHAR
LIFT GAS
MgO•CaS
$MgO•CaCO_3$
AIR
AIR
STEAM

Figure 1. Simplified flow diagram of clean gas at 14 atm pressure.

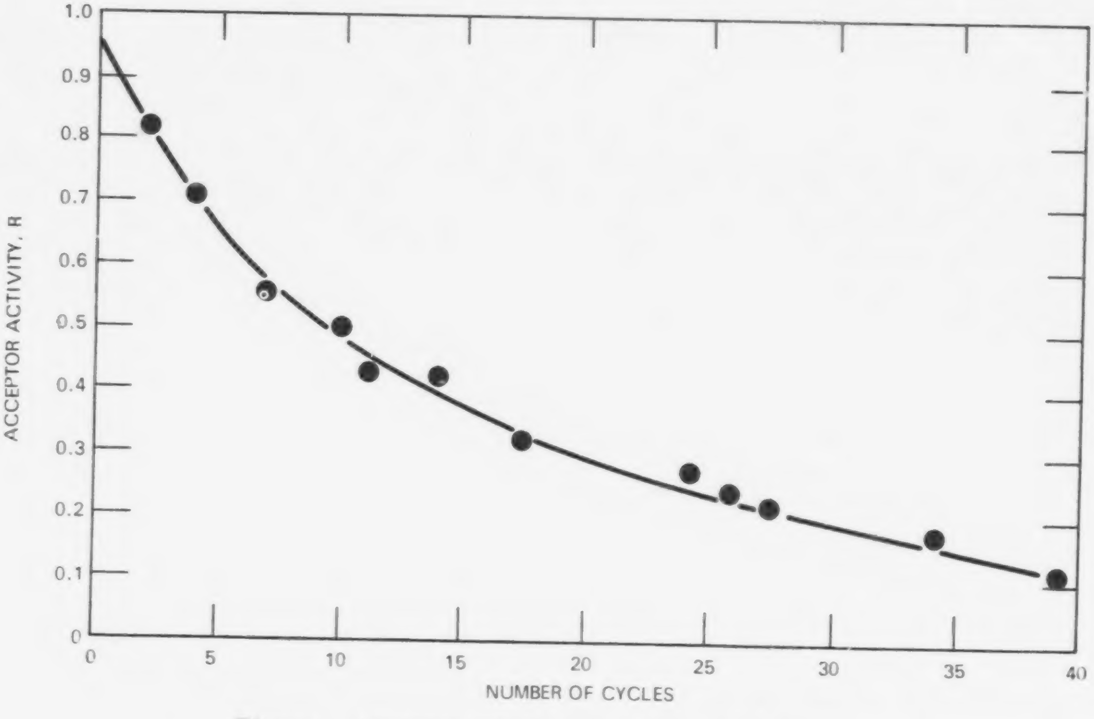

Figure 2. Acceptor activity versus number of cycles.

2. COAL DESULFURIZATION ASPECTS OF THE HYGAS PROCESS

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INTRODUCTION

One obvious alternative method for avoiding air pollution in the generation of energy from coal is to convert the coal to gas, desulfurize the gas, and utilize the low-pollution, sulfur-free gas to produce energy. Among the gasification processes now under study is the Hygas process being developed at the Institute of Gas Technology (IGT) under the joint sponsorship of the U.S. gas industry (via the American Gas Association) and the U.S. Government (via the U.S. Department of the Interior, Office of Coal Research). The former is seeking to secure a supplemental supply of gas equivalent to natural gas so that it will be assured of a future gas supply; the latter is vitally concerned with ensuring the availability of adequate energy supply and stimulating the utilization of our large coal reserves. This process not only converts coal to gas, but produces byproduct sulfur in elemental form.

The Hygas process has been under development at IGT since 1946. Until 1964, this work was supported almost entirely by the American Gas Association (AGA), at which time the U.S. Department of the Interior joined with the AGA in developing the process concept via a large-scale pilot-plant program. This pilot plant, now in the final construction and shakedown phase of development, is designed to produce about 1.5 million cu ft/day of gas from 80 tons of coal. It is hoped that an 80-million cu ft/day demonstration plant will be completed by 1974. Work, some of it still in progress, has been done in several smaller development units.

PROCESS DESCRIPTION

The Hygas process has been discussed in detail in a number of recent publications.[6,9] Figure 1 gives the basic steps. This process is operable with high- as well as low-rank coals, at a thermal efficiency of about 70 percent. However, it is at present necessary to pretreat agglomerating coals, such as the Eastern bituminous coals, before hydrogasification. Pretreating ensures that the coal will not agglomerate and hinder solids flow in the hydrogasifier. Nonagglomerating coals, such as those in the lignite and subbituminous class, do not require pretreatment. A substantial fraction of the sulfur may be removed, from some high sulfur coals, by pretreatment.

Hydrogasification is carried out in a two-stage fluidized-bed reactor. As now planned, coal or char is fed to the gasifier in a light oil slurry to avoid the use of lock hoppers. Small quantities of light oils are formed in the process; they vaporize in a drying zone at the top of the reactor, leave the reactor along with the raw product gas, and are recycled for reuse. Temperatures in the hydrogasifier range from about 1300°F to about 1800°F; reactor

Table 1. TYPICAL ANALYSES OF FEEDSTOCKS

	Coal seam				
	Pitts. No.8	Ohio No.6	Ill. No.6	Ind. No.6	W. Va. Block 5
Coal rank	HVBA	HVBB	HVBC	HVBC	HVBA
Mine	Ireland	Broken Aro	Crown	Minnehaha	Kanawha
County	Marshall	Coshocton	Montgomery	Sullivan	Kanawha
State	W. Va.	Ohio	Illinois	Indiana	W. Va.
Coals (before pretreatment)					
Ultimate analysis, wt%					
Carbon	67.90	74.40	70.10	73.20	79.00
Hydrogen	4.91	5.64	4.88	5.23	5.84
Nitrogen	1.18	1.23	1.10	1.29	1.34
Oxygen	6.38	9.64	9.82	8.73	6.04
Sulfur	4.38	3.36	4.00	2.34	0.80
Ash	15.30	5.73	10.10	9.21	6.98
Total	100.00	100.00	100.00	100.00	100.00
Proximate analysis, wt %					
Moisture	3.9	2.1	13.3	12.0	1.5
Volatile matter	32.6	39.2	33.0	32.7	35.7
Fixed carbon	48.8	53.1	44.9	47.2	55.9
Ash	14.7	5.6	8.8	8.1	6.9
Total	100.0	100.0	100.0	100.0	100.0
Chars (after pretreatment)					
Ultimate analysis, wt %					
Carbon	69.90	75.00	70.30	73.00	76.10
Hydrogen	3.09	4.18	3.58	3.43	3.97
Nitrogen	1.46	1.30	1.20	1.55	1.51
Oxygen	10.78	10.80	10.11	10.11	9.96
Sulfur	3.33	2.31	3.33	2.03	0.77
Ash	11.44	6.41	11.48	9.88	7.69
Total	100.00	100.00	100.00	100.00	100.00
Proximate analysis, wt %					
Moisture	1.4	1.4	1.4	0.5	0.6
Volatile matter	24.2	26.8	22.7	22.7	23.2
Fixed carbon	63.1	65.8	64.6	67.0	68.6
Ash	11.3	6.0	11.3	9.8	7.6
Total	100.0	100.0	100.0	100.0	100.0

USED IN HYGAS PROGRAM

Coal seam						
Sewell	Sewell	Pocahontas No.4	Laramie No. 3	--	--	--
MVB	LVB	LVB	Subbit.	Subbit.	Lignite	Lignite
Dante	Lockgelly No.2	Stotesbury No.10	Eagle	Colstrip	Glenharold	Savage
Nicholas	Fayette	Raleigh	Weld	--	Mercer	Sidney
W. Va.	W. Va.	W. Va.	Colorado	Montana	N. Dakota	Montana
83.20	87.40	85.10	75.10	67.9	66.0	64.8
5.04	4.85	4.20	2.56	4.52	4.37	4.17
1.52	1.52	1.12	1.33	1.01	0.56	0.95
5.58	2.86	2.81	15.90	17.86	20.00	21.22
0.62	0.60	0.73	0.38	0.81	0.99	0.68
4.04	2.77	6.04	4.73	7.90	8.08	8.18
100.00	100.00	100.00	100.00	100.00	100.00	100.00
1.4	1.1	0.7	3.7	3.0	5.0	4.3
29.4	20.6	16.1	35.2	36.6	40.8	39.3
65.2	75.6	77.2	56.5	52.7	46.5	48.6
4.0	2.7	6.0	4.6	7.7	7.7	7.8
100.0	100.0	100.0	100.0	100.0	100.0	100.0
80.20	85.10	85.30	73.10	--	--	--
3.65	3.32	2.23	4.27	--	--	--
1.59	1.49	0.84	1.40	--	--	--
9.85	7.84	5.24	15.62	--	--	--
0.59	0.58	0.59	0.27	--	--	--
4.12	2.67	5.80	5.34	--	--	--
100.00	100.00	100.00	100.00			
1.8	1.0	1.2	0.3	--	--	--
19.0	16.2	14.0	31.1			
75.2	80.2	79.1	63.3			
4.0	2.6	5.7	5.3	--	--	--
100.0	100.0	100.0	100.0			

pressures are up to about 1500 psig. Normally, sulfur removal is greatest in this processing step.

After the two stages of hydrogasification, the partially gasified char is gasified further with steam at 1800-1900°F in an electrothermal gasification stage. The hydrogen and carbon oxides produced in the electrothermal gasifier are fed directly to the hydrogasifier. The char from the electrothermal gasifier is then used as fuel for conventional power and steam generation. The sulfur content of this spent char is normally minimal.

The raw hydrogasifier effluent gases are water-quenched and the light oil and excess water are removed along with ammonia and coal fines. Next, the gas passes through an acid-gas-removal system that removes carbon dioxide and sulfur compounds - primarily hydrogen sulfide. After a final caustic and water wash, the gas is catalytically methanated to reduce the carbon monoxide content of the gas to less than 0.1 mole percent. The final gas has a sulfur level well below 1 ppm, a dew point of about -40°F, and a heating value of over 950 Btu/scf.

SULFUR REMOVAL

As was mentioned earlier, most of the sulfur is removed from the coal or char in three of the Hygas process steps: (1) coal pretreatment, (2) hydrogasification, and (3) electrothermal gasification. These steps are discussed in greater detail below.

Information on the amounts and types of sulfur compounds removed in these three stages is somewhat limited at this time. This limited information is the result of the research program on the Hygas process having been directed primarily toward developing a process for producing high-Btu gas from coal, with emphasis on hydrogasification, electrothermal gasification, and pretreatment–in that order. Sulfur removal aspects have been considered, but to a lesser degree. In addition, a wide range of coal feeds has been studied, which limits the amount of data on any one coal and makes it difficult to draw conclusions for some feedstocks.

Table 1 lists the coals that have been used as feedstocks thus far in this program. Most of the work has been with the Pittsburgh No. 8 seam, HVBA char; consequently, more is known about sulfur removal from this feed. For example, over 60 pilot-plant hydrogasification tests have been conducted on the Ireland mine coal, whereas only a few have been conducted with Illinois No. 6 and Ohio No. 6 coals.

Table 2 lists the pyritic and organic sulfur contents reported[12] by the U.S. Bureau of Mines for each type of coal studied. This data should only be considered as approximate, however, because we did not analyze in our laboratories samples of coals tested in the development unit.

Pretreatment

Agglomerating coals are pretreated by making them contact air in a fluidized bed at about 800°F. This process has been described in detail in earlier papers.[1,3] The sulfur removed is primarily as sulfur oxides, although there are undoubtedly small amounts of organic sulfur and other sulfur compounds present. The pretreater off gas could be treated with one of the many developing sulfur-oxide-removal processes to recover sulfur, or it could, after concentration of SO_2, be combined with the hydrogen-sulfide-containing gases removed from the hydrogasifier effluent and sent to a Claus plant for elemental sulfur production. The large pilot plant now being completed will contain a Claus-type plant.

Figure 2 shows the results of typical pretreatment tests for the coals listed in Tables 1 and 2. Although only a limited amount of data is available and the data exhibits considerable scatter, it is clear that sulfur removal in pretreatment increases with increases in total sulfur content. Accurate measurement of the fraction of sulfur removed is difficult, especially for low-sulfur coals, because of the small differences in the sulfur contents of most of the coals and chars. Inaccuracies in material balances, variations in pretreatment levels, and sampling errors all contribute to the overall error in this measurement.

Figure 3 shows the effect of organic sulfur content of the coal on sulfur removal. Apparently pyritic sulfur is more easily removed from the bituminous coals than organic sulfur. The higher sulfur removal measured for the lower rank subbituminous coal is not considered significant in view of its very low total sulfur content.

We expect that more information will be obtained from the results of the operation of the large pilot plant now being completed. In general, however, we can expect to remove from 20 to 40 percent of the sulfur present in bituminous coals in the pretreatment step.

Table 2. TYPICAL SULFUR BREAKDOWNS REPORTED FOR TYPES OF COALS TESTED[a]

Coal	Type	Pyritic sulfur wt %[b]	Organic sulfur wt %[b]	Total sulfur wt %[b]
Pittsburgh No. 8	HVBA	2.20	2.27	4.48
Ohio No. 6	HVBB	1.80	1.61	3.48
Illinois No. 6	HVBC	2.84	2.17	5.09
Indiana No. 6	HVBC	1.15	0.87	2.06
W. Va. Block 5	HVBA	0.05	0.55	0.61
W. Va. Sewell	MVB	0.06	0.51	0.57
W. Va. Sewell	LVB	0.04	0.50	0.55
Pocahontas No. 4	LVB	0.09	0.51	0.60
Laramie No. 3	Subbit.	0.06	0.22	0.28
Montana	Subbit.	0.39	0.41	0.82
N. Dakota	Lignite	0.02	0.85	0.88
Montana	Lignite	0.37	0.36	0.77

[a] Reported by U. S. Bureau of Mines, IC 8301.[12]

[b] Dry basis.

Hydrogasification

More data is available on sulfur removal during hydrogasification than during pretreatment because this program has been concerned primarily with the hydrogasification step. Details on this step have been published previously.[7,8] When coals and chars are hydrogasified with either hydrogen-steam mixtures or synthesis gas, sulfur is removed primarily as hydrogen sulfide and carbonyl sulfide, which can be removed from the hydrogasifier effluent by a number of well-developed processes. One such approach, for example, is to scrub out H_2S, COS, and CO_2 by the hot carbonate process and then convert the H_2S to elemental sulfur in a Claus plant, as was discussed above. COS is hydrolyzed to H_2S in the gas-cleanup operation. The Claus plant feed could contain (on a dry basis) from about 3 mole percent (for lignite feeds) to about 32 mole percent (for bituminous coal chars and high-hydrogen-content feed gases to the hydrogasification section), if the pretreater off gas were combined with the H_2S, and CO_2 were removed from the hydrogasifier effluent.

Figure 4 shows that, as in the pretreatment test data, considerable scatter exists in the sulfur removal data for hydrogasification. The major trend observed is that the degree of char sulfur removal varies directly with the MAF char-gasification level. In contrast to the pretreatment results, sulfur is removed more readily from chars from coals with the highest organic-sulfur contents. The lignite feeds, which were not pretreated and which hydrogasify much more readily than higher rank feeds, gave results comparable with the other feeds. In addition, the results with North Dakota lignite compare favorably with those for Montana lignite, even though their organic/pyritic sulfur ratios differ markedly. The West Virginia coals, all of which have a high organic/pyritic sulfur ratio, allowed even higher sulfur removal. This is just the opposite of the pretreatment results, but is in agreement with the coal hydrodesulfurization work of Vestal and Johnston.[11]

The large amount of scatter in the data shown in Figure 4 suggested that other variables (e.g., temperature, pressure, and gas composition) might also be important. The only approach available at that point seemed to be regression analysis becuase the test program to date had been concerned with hydrogasification variables, not with those variables influencing sulfur removal. We attempted, therefore, to correlate the results with Ireland-mine coal char (the majority of testing was done with this feed material) using multiple linear regression analysis.

There were three main sets of data: those using hydrogen-steam feed-gas mixtures, those using hydrogen-methane steam, and those using synthesis gas. The results of the regression analysis showed that the only statistically significant variable for the synthesis gas and hydrogen-methane-steam feed-gas runs was temperature. The hydrogen-steam run results showed a dependence on both MAF-char gasification and feed-gas/char ratio. We then decided that these results were subject to too great an experimental error for conventional techniques. We did try to force-fit the results by standard techniques; Figure 5 shows that the predicted results agreed, in general, quite well with the experimental data. It appears that further work will be required to establish precisely the variables affecting sulfur removal. It is possible, however, to estimate sulfur removal from the coals tested to within about ±10 percent. Using this information, we can expect to remove anywhere from 40 percent to essentially all of the feed-char sulfur content in the hydrogasification step.

Electrothermal Gasification

At the present time, less data is available from our electrothermal gasification program. Again, details of this program have been published elsewhere.[4,5] Because the gas produced in the electrothermal gasifier passes directly into the hydrogasifier, it is important to be able to predict its sulfur content. Any sulfur compounds evolved here will ultimately end up in the hydrogasifier effluent.

The data available correlates better for these tests (Figure 6) than for those discussed previously. It should be pointed out that the variable is "char" gasified (in the electrothermal gasifier), not "MAF-char" gasified. Also, the sulfur removal data is based on the sulfur content of the feed to the electrothermal gasifier; this feed has had most of its sulfur removed in the two previous processing steps–pretreatment and hydrogasification.

The closer agreement of the data from the various feeds tested may stem from their having similar properties: they are all highly gasified materials. Earlier work[2] has shown that the reactivities of coals, ranging in rank from lignite to anthracite, approach one another at high levels of gasification.

Economics of Sulfur Removal

Figure 7 shows a typical breakdown of sulfur removal from the coal in each stage of the Hygas process. It is difficult to extract the true cost of sulfur removal from available data because of the uncertainties in the future value of sulfur. It is also difficult to separate sulfur-removal costs from the total costs of gas purification. However, if elemental sulfur could be sold for $25/long ton, the Hygas process economics might be more favorable for higher sulfur coals; the cost of sulfur removal would then be less than the sulfur byproduct credit. For low-sulfur coals and a lower sulfur price, the reverse would be true. These figures assume a final gas cost of 5-6¢/therm (based on AGA accounting procedures), a coal cost of 16¢/million Btu, and present economic conditions in the United States. A detailed study[10] on sulfur recovery in the manufacture of pipeline gas from coal has recently been presented.

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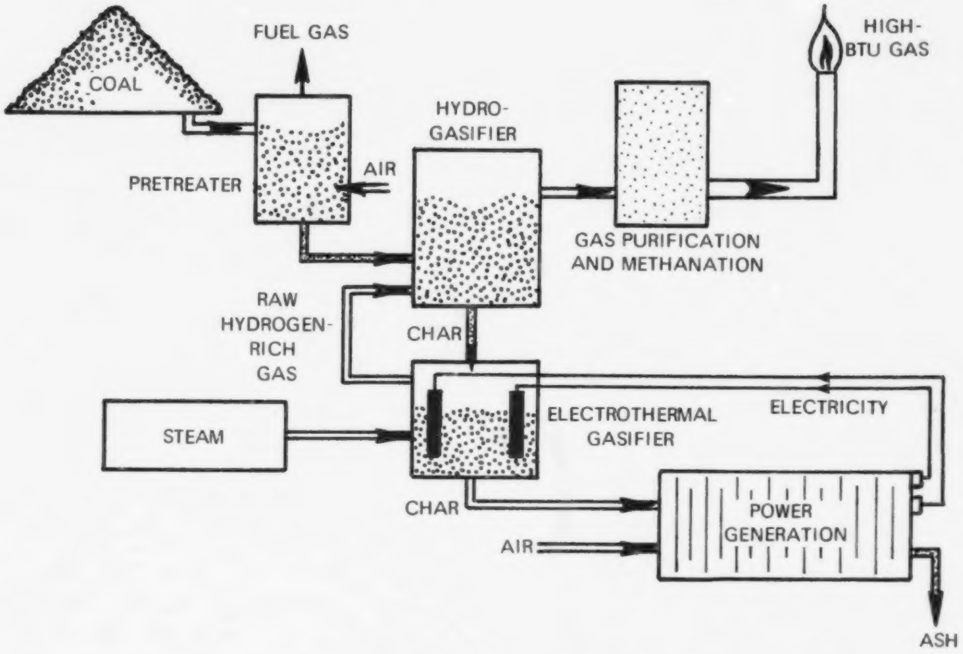
FUEL GAS
COAL
PRETREATER
AIR
HYDRO-GASIFIER
HIGH-BTU GAS
GAS PURIFICATION AND METHANATION
RAW HYDROGEN-RICH GAS
CHAR
STEAM
ELECTROTHERMAL GASIFIER
ELECTRICITY
CHAR
AIR
POWER GENERATION
ASH

Figure 1. Schematic flow sheet of Hygas process.

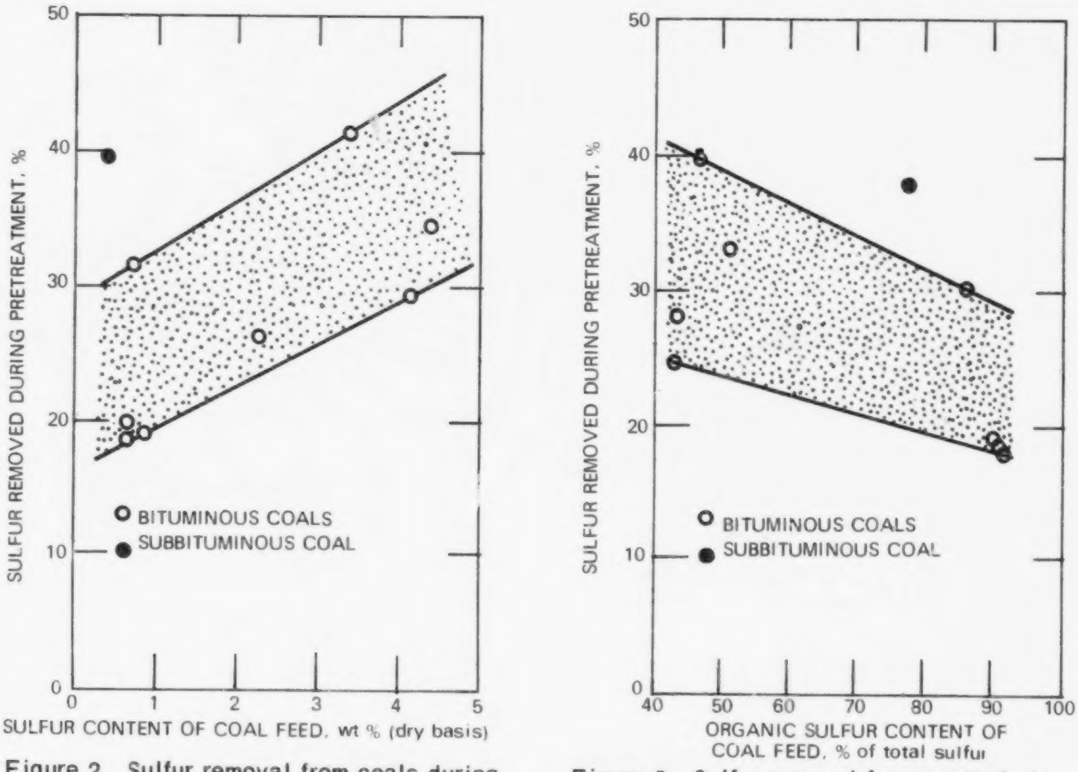

Figure 2. Sulfur removal from coals during pretreatment increases with increasing sulfur content.

Figure 3. Sulfur removal from coals during pretreatment decreases with increasing organic sulfur content.

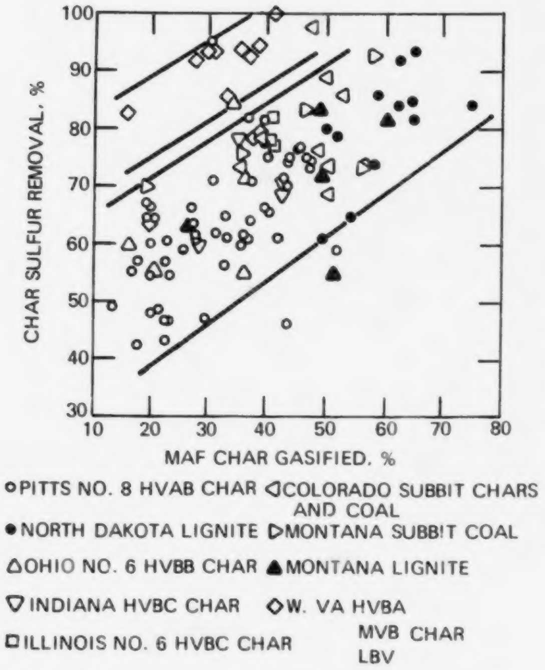

Figure 4. Char sulfur removal increases with char gasification in hydrogasifier.

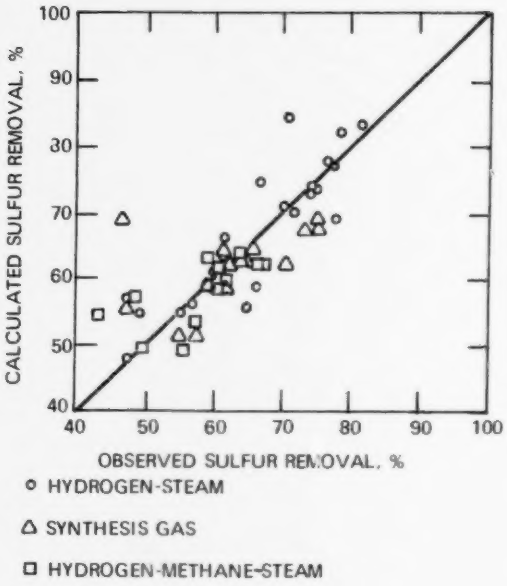

Figure 5. Agreement between measured sulfur removal data and forced-fit data.

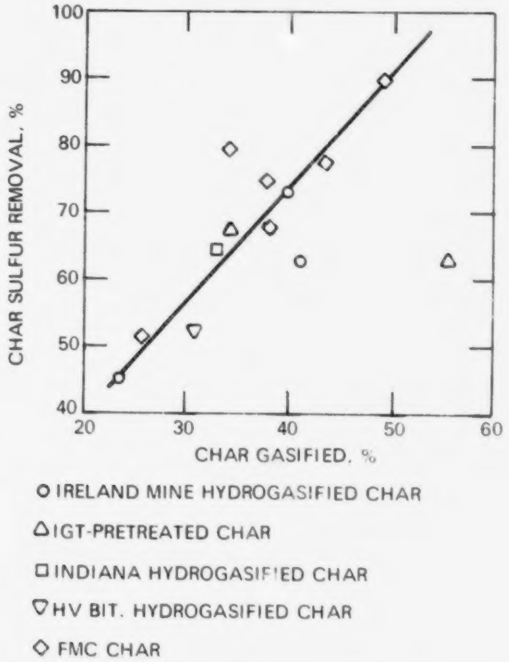

Figure 6. Char sulfur removal in electrothermal gasifier increases with increasing char gasification.

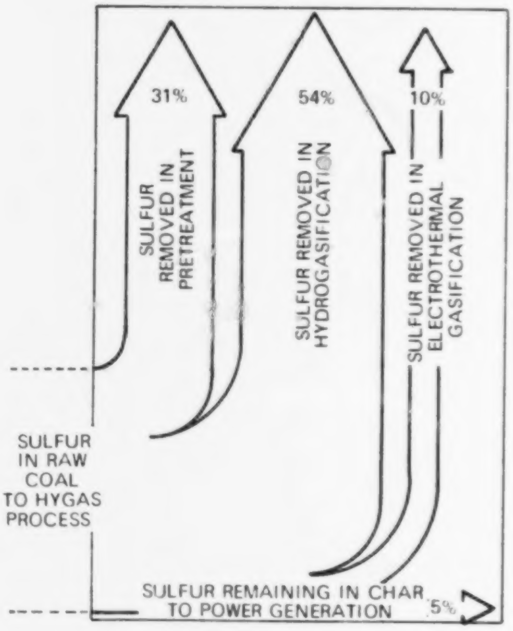
31%
SULFUR REMOVED IN PRETREATMENT
54%
SULFUR REMOVED IN HYDROGASIFICATION
10%
SULFUR REMOVED IN ELECTROTHERMAL GASIFICATION
SULFUR IN RAW COAL TO HYGAS PROCESS
SULFUR REMAINING IN CHAR TO POWER GENERATION
5%

Figure 7. Typical sulfur removal in each stage of the Hygas process.

3. STEAM-OXYGEN GASIFICATION OF VARIOUS U. S. COALS

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U. S. Bureau of Mines

ABSTRACT

The Bruceton process for making a supplemental natural gas from coal consists of gasification, purification, and methanation steps. The advantages of the system are that caking coals can be used in the fluid-bed gasifier, and more than half of the methane can be made in the gasifier instead of in the methanator. Furthermore, the final product contains no sulfur and has a heating value exceeding 900 Btu/cu ft.

INTRODUCTION

The need for a supplemental natural gas is becoming more evident each year. The reserves-to-production ratio, now about 14:1, has been declining. If the cost of natural gas continues its present upward trend, a substitute natural gas will be able to compete with the natural product in the immediate future.

One source of this substitute gas is to make it from coal because of its widespread abundance and low cost. The steam-oxygen process for making supplemental gas from coal not only makes the product inexpensively, but also decreases the sulfur pollution to a tolerable level. The process has been previously reported in the literature.[1,2,3]

All of the work described here was done at the Bruceton, Pa., laboratories of the Pittsburgh Energy Research Center of the Bureau of Mines.

THE OVERALL PROCESS

The Bruceton process for making a supplemental natural gas is shown schematically in Figure 1. The main components are: (1) the gasifier; (2) the shift converter, which changes the gas ratio from approximately $2H_2$:1CO to $3H_2$:1CO; (3) the purification system, which removes most of the CO_2 and all of the sulfur compounds from the gas; and (4) the catalytic methanator, in which $3H_2$ + 1CO reacts to form $CH_4 + H_2O$. Because the shift and purification systems are commercial, research at the Bureau is concentrated on the gasification and the methanation steps. This paper will concentrate on the gasification step; it should be emphasized, however, that the gas is 400-500 Btu/cu ft before methanation and afterwards should be of pipeline quality—at least 900 Btu/cu ft. Because of the nickel catalyst used in the methanation step, the sulfur level cannot exceed 1 ppm; therefore, the final product may be considered sulfur-free.

The gasifier is shown schematically in Figure 2 and photographically in Figure 3.

The Bructon gasifier combines the operations of pretreatment, carbonization, and gasification in one reactor. The coal, 70 percent through 200 mesh, is dropped through the pretreater with oxygen and steam (or CO_2) at 400°C, where it is rendered noncaking. The decaked coal then falls into the carbonization zone and finally into the gasification zone, where it is gasified with steam plus oxygen at 900-1000°C. The gasifier is operated at 40 atmospheres pressure.

Advantages of the Bruceton gasifier are: (1) it can handle both caking and noncaking coals, (2) gases from pretreatment add to the product gas from the gasifier, and (3) it makes a purified product gas with a very high (20-30 percent) methane content (the balance is H_2 - CO).

This high methane content means the volume of gas is less than if it were H_2 + CO only. Thus, the size of all process vessels and lines downstream from the gasifier are reduced as much as 30-40 percent; the final methane reactor is reduced about 50 percent from the size it would need to be if only H_2 + CO were in the feed gas.

The Bruceton process has been estimated to have an overall thermal efficiency of 63 percent. The selling price for the product gas has been estimated at 54¢/M cu ft, a price that is competitive with that of other coal-to-gas processes.[4]

Table 1 shows the results of selected tests with four coals. The free-swelling index (FSI) is used to indicate the caking or coking property of each coal. Pittsburgh seam, a highly caking coal, has a maximum FSI of 8 to 9; Illinois No. 6, a weakly caking coal, has an FSI of about 4.5; and North Dakota lignite and Montana subbituminous coal have an FSI of 0 (i.e., they are noncaking).

Table 1. RESULTS OF TESTS WITH VARIOUS COALS GASIFIED AT 40 ATMOSPHERES

Test	Type coal				Goals
	Pittsburgh Seam	Illinois No. 6	North Dakota Lignite	Montana Subbit.	
Free-swelling index	8 to 9	4.5	NC[a]	NC[a]	-
Carbon conversion, %	67	72	88	71	65
Product gas, H_2+CO +CH_4, scf/lb MAF coal	17	18	25	16	17
CH_4, scf/lb MAF	3.9	3.7	4.2	3.0	4.7
Gas analysis, %					
H_2	34	36	35	38	-
CO	19	19	23	11	-
CH_4	16	14	15	11	-
CO_2	32	31	29	40	-
Tar, % feed	2.9	3.4	2.0	3.3	-

[a]NC = Noncaking.

The carbon conversion for the tests shown in Table 1 ranges from 67-88 percent; our goal is 65 percent. This degree of conversion permits the complete utilization of carbon in the overall process because the char and tar can be burned to raise the steam needed for compression and other steps in the process. Other goals required to make the process economical are listed in Table 1. It is desirable to make about 17 scf of product gas per lb MAF coal because this results in a final high-Btu gas volume of 8 to 9 scf per lb MAF coal. We want to make 4.7 scf CH_4 per lb MAF coal, but are slightly low on this goal. The gas analyses are similar even though different coals are used.

Table 2 shows that the gas leaving the gasifier contains most of the sulfur in the coal. Some sulfur is left in the char and some in the tar; however, both will be burned.

It is desirable—even necessary—that the sulfur content of both the char and tar be below 1 percent so they can be burned without discharging unacceptable amounts of SO_x into the air. Table 3 shows that this limitation is achieved with all coals tested except Illinois No. 6. This coal, therefore, and similar high-sulfur coals may present a problem. If the carbon conversion is increased to lower the

Table 2. DISTRIBUTION OF SULFUR DURING GASIFICATION[a]

Sulfur distribution, lb	Type coal Pittsburgh Seam	 Illinois No. 6	 N. Dakota Lignite	 Montana Subbit.
Coal	26.0	78.0	16.0	28.0
Char	3.1	11.2	4.4	4.4
Tar	0.5	1.1	0.4	0.3
Gas[b]	22.4	65.7	11.2	23.3
Total S in gas, %	86	84	70	84

[a] Basis: per ton of coal feed to gasifier.

[b] By difference.

Table 3. SULFUR CONTENT OF COALS AND PRODUCTS

Sulfur content, wt %	Type coal Pittsburgh Seam	 Illinois No. 6	 N. Dakota Lignite	 Montana Subbit.
Coal	1.3	3.9	0.6	1.4
Char	0.5	1.4	0.6	0.9
Tar	0.8	1.7	1.0	0.5
Gas	0	0	0	0

sulfur in the char the amount of char is decreased. This means low-sulfur supplemental coal must be burned to raise the steam required for the overall process. Another possibility is that the coal could be washed to achieve a lower level of sulfur before gasification.

CONCLUSION

The high-Btu gas made in the Bruceton process is low enough in sulfur that it causes no problem with air pollution requirements. The chars and tars from most coals can be burned to raise steam and can be made so the total sulfur in the feed to the power plant is less than 1 percent. Some high-sulfur coals may present a problem, however, because the sulfur in the char and tar may be too high. More research will be required to resolve this problem.

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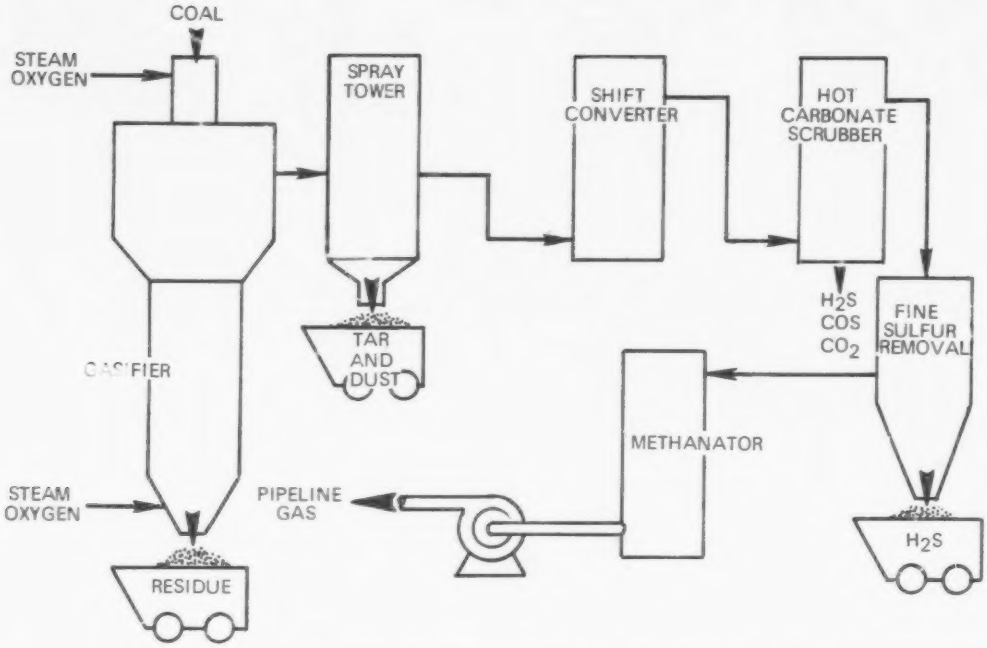
COAL
STEAM OXYGEN
SPRAY TOWER
SHIFT CONVERTER
HOT CARBONATE SCRUBBER
GASIFIER
TAR AND DUST
H_2S COS CO_2
FINE SULFUR REMOVAL
METHANATOR
STEAM OXYGEN
PIPELINE GAS
H_2S
RESIDUE

Figure 1. Flow diagram of system used to make high-Btu gas from coal.

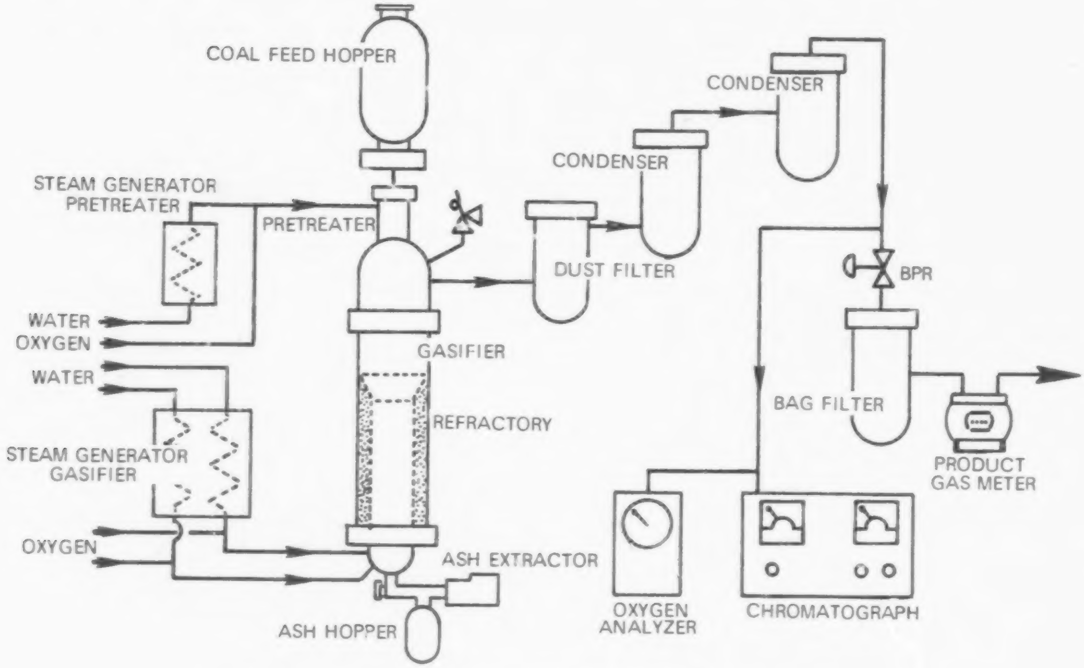
COAL FEED HOPPER
CONDENSER
CONDENSER
STEAM GENERATOR PRETREATER
PRETREATER
DUST FILTER
BPR
WATER
OXYGEN
WATER
GASIFIER
REFRACTORY
BAG FILTER
STEAM GENERATOR GASIFIER
PRODUCT GAS METER
OXYGEN
ASH EXTRACTOR
ASH HOPPER
OXYGEN ANALYZER
CHROMATOGRAPH

Figure 2. Diagram of a 40-atmosphere fluid-bed gasifier.

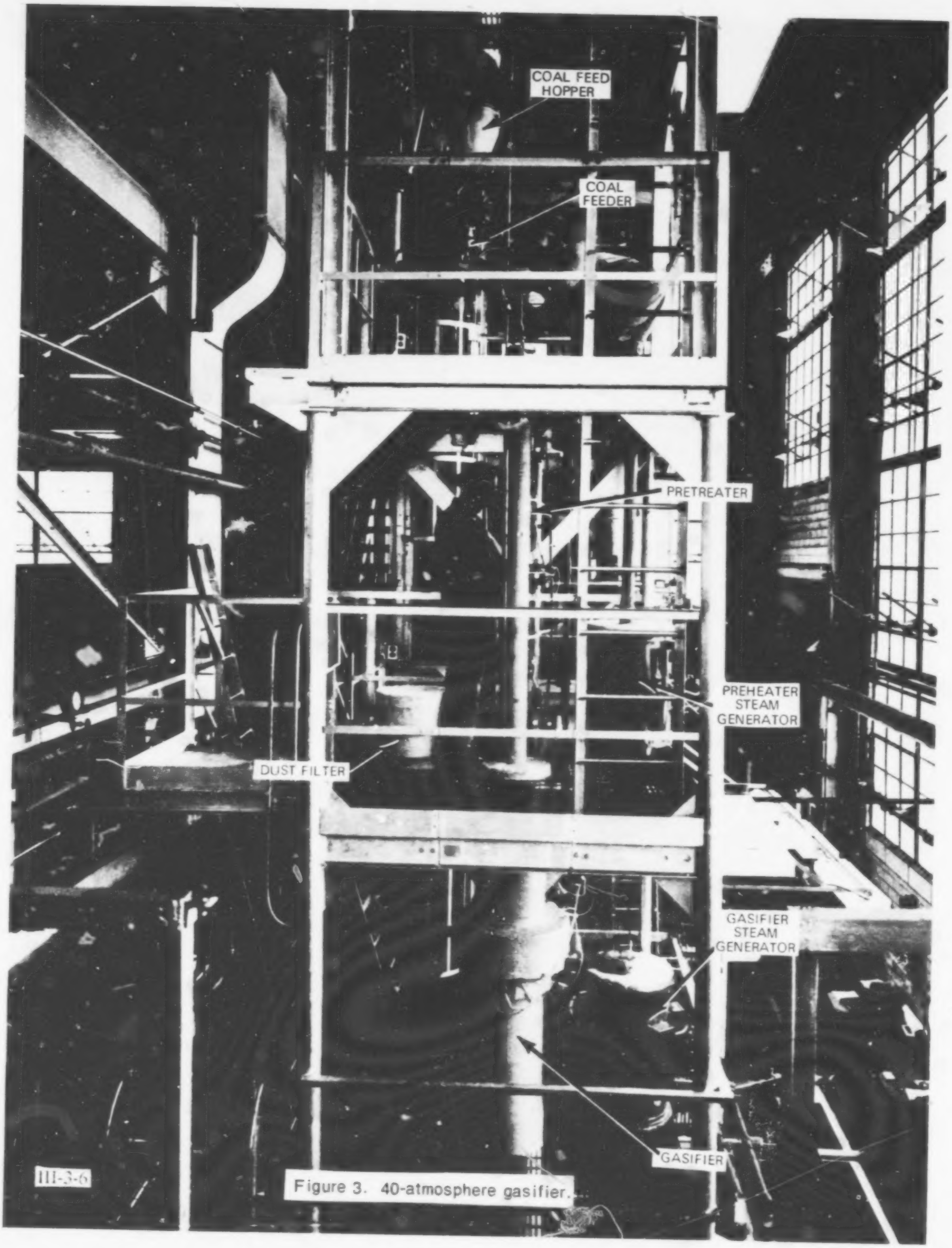
COAL FEED HOPPER
COAL FEEDER
PRETREATER
PREHEATER STEAM GENERATOR
DUST FILTER
GASIFIER STEAM GENERATOR
GASIFIER

Figure 3. 40-atmosphere gasifier.

4. DESULFURIZED FUEL FROM COAL BY INPLANT GASIFICATION

E. K. DIEHL AND R. A. GLENN

Bituminous Coal Research, Inc.

ABSTRACT

The growing demand for electrical power will continue to draw on fossil fuels as a source of energy. At the same time, the need to control and reduce environmental pollution will dictate the consumption of fossil fuels in a manner that will minimize their contribution to atmospheric pollution.

For coal, a solution lies in the conversion of the solid fuel into a clean, sulfur-free fuel gas. This fuel gas can be subsequently utilized in a variety of downstream systems, many of which promise overall improvements over conventional power-generating systems.

Bituminous Coal Research, Inc. (BCR) is developing, for the Office of Coal Research, U.S. Department of the Interior, a two-stage, super-pressure, oxygen-blown coal gasifier for the production of pipeline gas. Results obtained thus far have led to the conclusion that a similar air-blown system would be advantageous for the production of a fuel gas suitable for desulfurization and pollution-free combustion in power plants.

This paper describes the BCR gasifier and suggests several alternate uses of the clean-fuel gas so that a major reduction in atmospheric pollution can be realized with continued use of coal in the growing energy market.

INTRODUCTION

As a result of the increased concern over environmental pollution, strict limitations and control requirements are being imposed on processes emitting atmospheric contaminants. This is especially true of fossil-fuel-fired electric generating stations that may emit fly ash and/or sulfur oxides. Flue gases can be processed to minimize fly ash emission at reasonable costs. Currently, the development of equally satisfactory methods for the control of sulfur oxide emission is the principal objective of major research programs being conducted by both Government and industry.

The electric power generating industry, in particular, finds itself on the horns of a dilemma. The demand for electricity within our society continues to grow at a minimum rate of 7 percent each year, with a doubling time of about 10 years; in 1968 and 1969, the growth rate was 9 percent. Generating capacity to meet this demand is being constructed, planned, and projected on a scale never before experienced.

Although there is a wide difference of opinion concerning how this mushrooming generating capacity will be fueled, there is general agreement that for the next several decades coal will remain a major source of energy fuel. Thus the dilemma for the industry: huge coal resources will continue to be tapped to keep pace with the electric power demand, but environmental pollution control will dictate their consumption in a manner that will assure minimum atmospheric contamination.

While there may still be some minor gains to be realized in conventional fuel-to-electricity cycles, research is rapidly shifting toward the development of new unconventional concepts. Basic to the development of these new concepts is the realization that they must include features that reduce or eliminate environmental pollution problems. Further, new concepts, in order to be accepted and adopted by the power generation industry, must be economically competitive with conventional systems equipped with pollution control devices.

In the recent years, terms such as magnetohydrodynamics, electrogasdynamics, advanced-cycle power systems, fuel cells, fluidized-bed combustion, and gasification have become commonplace wherever technical people gather to contemplate the future of power generation. It is obvious that unconventional concepts are receiving serious attention, and that they will, in time, take their place in the total power generation picture.

For coal-fired installations, a promising approach is the conversion of the coal to sulfur- and dust-free gas prior to its ultimate use as an energy fuel. The careful selection of specific operating parameters and the application of new technology can lead to the production of gas having a gross heating value ranging from about 200 Btu/scf to as high as 950 Btu/scf.

Such a system holds promise as a means of generating power not only without air pollution, but also with decreased overall cost, especially if recovered sulfur can be sold as a credit against the cost of the process. In addition, a reduction of nitric oxide emission is conceivable because of the more precise combustion control possible with gaseous fuels.

GAS PRODUCERS

The only commercial fixed-bed process for the gasification of coal at elevated pressure available today fulfills the requirement for gasifying coal with high thermal efficiency. However, this process is not suitable for the gasification of highly caking coals and is limited to the use of lump fuel. The permissible gas-flow velocity limits the size of the individual gasifiers, as well, and for a large utility power plant, costly multiple units would be required. The production of tar inherent in this process also adds to the process cost because purification of both the liquid and the gaseous effluents then becomes necessary.

Coal-gasification processes have also been developed, using fluidized beds. The Winkler process, operating at atmospheric pressure, has been used in large units burning predominantly brown coal. Operation of the process at elevated pressure has been carried out in pilot plants only, and with only noncaking fuels. Fluidized-bed gasification processes leave a residue with a high carbon content that requires separate utilization and thus complicates the process economics. The operability of fluidized beds at elevated pressure using caking coals has not, moreover, been established. This system requires development work on a large scale, and may impose limitations on such a process.

The simplest process is the gasification of coal in the entrained state. Very large units can be built and complete carbon conversion assured by operating under slagging conditions. These processes use coal at any rank, regardless of caking properties. Many commercial plants, operating at atmospheric pressure, have been built to use peat, brown coal, and bituminous coals as raw material. Operability of this type of process at elevated pressure has been proved in pilot plants.

BCR Two-Stage Gasifier

BCR is developing, under sponsorship of the Office of Coal Research, a two-stage process for the gasification of coal for the production of high-Btu pipeline gas.[1,2,3] The process combines the advantages–high gas yield and low exit temperature–of the more complex fixed- and fluidized-bed processes with the simplicity of entrained processes and their ability to use any coal.

The basic component of the process under development by BCR is the two-stage gasifier (Figure 1). Fresh coal and steam are introduced into the upper section (Stage 2) of the gasifier, where the fresh coal is contacted and entrained by a rising stream of hot synthesis gas produced in the lower section (Stage 1). The fresh coal is rapidly heated and partially gasified to methane and additional synthesis gas in Stage 2. Residual char, swept out of the gasifier along with the raw gas, is separated, then returned to Stage 1. Here the char is completely gasified under slagging conditions to produce both the synthesis gas and the heat required in Stage 2.

For the production of pipeline gas, Stage 1 is blown with oxygen and steam, and the product gas from Stage 2 undergoes purification, partial shift reaction, and catalytic methanation to raise the heating value above 900 Btu-/scf. The gasifier is operated at about 1000 psi.

Experimental results obtained so far in the development of the two-stage, super-pressure process have led to the conclusion that, if blown with air rather than with oxygen, the two-stage principle would also be advantageous for the production of a fuel gas suitable for desulfurization and pollution-free combustion in power plants. Thus, the air-blown gasifier could be used to generate fuel gas from which sulfur, as well as particulates, could be removed by existing cleanup systems. The resultant clean, sulfur-free gas could then be utilized in a variety of power-generating systems.

Air-Blown Gasifier

Using information generated in the pipeline gas project, studies have been made to verify the technical feasibility of using the two-stage gasifier to supply a fuel gas. Figure 2 is one example of a flow diagram and material balance for a conceptual fuel-gas system. A basis of 100 lb of moisture and ash-free coal is used for the material balance, the gasifier is operated at 20 atmospheres, and the gasifier outlet temperature is 1700° F.

Steam and air are preheated to 1700°F. For the purpose of the example, it is assumed that progress in materials development and the small pressure differential between products and feeds can make possible this preheat step.

Table 1 gives analyses of the feed coal and resultant fuel gas; Table 2 shows a simplified heat balance.

Mathematical Model

The material and heat balances for the example system were estimated using a mathematical model developed at BCR.[4] Figure 3 shows the general scheme of the model. In Stage 1, all of the materials are mixed thoroughly and the makeup char is gasified. The resulting mixture comes to water-gas shift equilibrium at the Stage 1 temperature that results from the Stage 1 energy balance based on an assigned heat loss. This gas moves to Stage 2 and contacts the coal, where the following reactions occur:

$$C + 2H_2 \rightarrow CH_4 \qquad (1)$$

$$C + H_2O \rightarrow CO + H_2 \qquad (2)$$

In the gasifier model, several general assumptions were made pertaining to the thermodynamics of the process:

1. The water-gas shift equilibrium adequately describes the gas composition from Stages 1 and 2.
2. The enthalpies of mixing of the gases are zero.
3. All gas fugacities and activity coefficients are unity.
4. The effect of pressure on the heat of reaction is minor.

Table 1. ANALYSES OF COAL AND PRODUCT GAS

Coal analysis		Fuel-gas analysis	
Constituent	Percent	Constituent	Mol percent
Carbon	70.1	CH_4	3.3
Hydrogen	4.9	CO	22.7
Oxygen	6.6	CO_2	7.0
Nitrogen	1.4	H_2	24.9
Sulfur	3.0	H_2O	10.9
Ash	12.7	N_2	30.7
Moisture	1.3	H_2S	0.5
Total	100.0	Total	100.0
H.V., gross	12,800 Btu/lb	H.V., gross, dry	210 Btu/scf
H.V., net	12,300 Btu/lb		
H.V., net. MAF coal	14,350 Btu/lb		

Table 2. HEAT BALANCE, AIR-BLOWN GASIFIER[a]

Heat input		Heat output	
Constituent	M Btu	Constituent	M Btu
Coal, H.V., net	1435.2	Gas, H.V., net	1332.1
Coal, sensible heat	22.8	Gas, sensible heat	263.4
Steam	87.0	Slag	12.4
Air	90.8	Char	58.6
Char	34.4	Loss	3.7
Total	1670.2	Total	1670.2

[a]Basis: 100 lb MAF coal; 20 atm; exit gas at 1700°F; steam/coal ratio 1.06.

Kinetic equations for methane yield and carbon oxide formation, together with appropriate material and energy balances, were programmed for a digital computer. The resultant simulation was used to obtain both the foregoing balances and the relationships that follow.

Effects of Major Operating Variables

Using the mathematical model, the system was tested for the effects of changes in steam-coal ratio, operating pressure, and gasifier outlet temperature. The resulting influence of these variables on the heating value of the product gas, the producer-gas volume, and the thermal efficiency of the system is shown in Figures 4, 5, and 6.

The strongest effect is that of the gasifier outlet temperature of the heating value of the product gas. If a higher temperature is required by a certain fuel-utilization system, the heating value of the gas can be increased by changing one or more of the other operating variables.

Although the system study presented here is by no means exhaustive, it shows the operational flexibility of the gasifier. Thus, depending upon downstream use of the fuel, such operating variables as steam-coal ratio, temperature, and pressure may be selected to optimize costs, to provide for convenience of operation, or to reflect downstream needs.

Sulfur Removal

Because sulfur appears in the gasifier product gas as H_2S, its removal is not a major problem. Reduction of both gas temperature and pressure, if required, can be accomplished without undue penalty to the system. Such processes as the Benfield and iron oxide fit the system quite well.

Current Status

For the further development of an air-blown two-stage gasifier for commercial-scale use, the next step is considered to be the operation of the system at the pilot-plant level. A technical evaluation and cost estimate of a 5-ton/hr pilot plant was recently completed with the assistance of Koppers Company, Inc., as an outside engineering subcontractor. In addition to the basic gasifier, the pilot plant would include coal preparation and handling facilities, gas desulfurization units, effluent waste treatment, and other general facilities needed for the conversion of coal to sulfur-free, low-Btu fuel gas.

Cost evaluation is continuing. It has been expanded in scope to include an estimate of the cost of a commercial plant as well as the ultimate cost of the clean fuel gas.

FUEL GAS UTILIZATION

The clean, desulfurized, high-pressure fuel gas could be utilized through several alternate routes. Perhaps the least efficient use of the gasifier product would be its combustion in a conventional steam boiler. Although the ash-free quality of the gas would eliminate the need for boiler-cleaning equipment, and freedom from sulfur would solve a major air pollution problem, it is doubtful if these benefits would ordinarily offset the coal conversion process. It is conceivable, nevertheless, that gasification of available coal could be the best solution in a particularly vital energy situation. Low-pressure gasification could, for example, supply fuel gas directly to a conventional boiler. If more efficient gasification would be attained at higher pressures, the gasifier product gas could be expanded through a gas turbine before being burned.

A more efficient use of the gasifier product would be its combustion in a supercharged boiler. With the addition of a topping gas turbine to utilize the work available in the high-pressure flue gas, the system shows some advantages over conventional operation. The cycle efficiency can be improved if, in addition to driving a combustion air compressor, the topping turbine is coupled to a generator. While steam remains the major generating fluid, the gas turbine can add to the net generation and at the same time eliminate other fan power requirements.

Although present-day supercharged boilers do not operate much above 5-7 atmospheres, it is conceivable that higher pressures could be utilized. Little economic gain, however, can be realized above 7 atmospheres. Nevertheless, Figure 5 shows that the gasifier can be operated at 5 atmospheres without appreciably affecting gas quality or thermal efficiency.

Combined Cycles

Over the past several years, many examples of conceptual combined cycles have been studied. A recent paper[5] by Robson and Giramonti evaluates a number of combined cycles and establishes a timetable based on gas turbine development. Known by various names, most cycles have in common the major generation of power by a gas turbine, which then exhausts into a steam boiler at essentially atmospheric pressure. Net generation includes that obtained from a steam turbine.

There are many variations on the combined-cycle concept. Some publicized cycles limit themselves to current commercial gas-turbine inlet temperatures, approximately 1800°F. Others, however, expressing firm belief in advancing gas turbine technology, assume higher temperatures in order to demonstrate the potential of the combined cycle.

Direct coal-fired gas turbines, the object of some past and current research, are faced with a major problem area: erosion and corrosion from coal ash and certain ash constitutents. The coal gasifier, on the other hand, because it produces a clean product gas, is an excellent means for using coal as a source of energy for the combined cycle.

Figure 7 shows a conceptual 1000-MW combined-cycle power generating system. This system, conceived[6] by Curtiss-Wright Corporation, uses the BCR two-stage gasifier to generate fuel gas at 22 atmospheres and 2000°F. (Note that the cycle incorporates liquid-metal heat exchange to reclaim the sensible heat in the product gas before it is cleaned.) Further, the gas turbine inlet temperature is shown as 3196°F. While both of these innovations are not considered to be currently commercial, they are not believed to be beyond grasp. Sufficient incentive for their development may be fostered by the promise that the combined cycle holds.

The Curtiss-Wright plan shows that a system heat rate of 6740 Btu/KWH can be obtained using 12,800-Btu/lb coal. Although this gain in cycle efficiency may be somewhat optimistic, because it assumes successful development of the innovations mentioned above, it does indicate that the combined-cycle concept deserves further attention.

With gasification and gas cleanup at elevated pressures, the size and possibly the cost of the gas producer would be significantly less than with atmospheric pressure systems. In addition, the power cycle's increased efficiency, coupled with the air pollution control aspects of the system, should help to supply a solution to the growing need for clean power.

Engineering development of the BCR two-stage gasifier for the production of pipeline gas is entering the pilot-plant stage. Current projections indicate that a commercial plant can become a reality in about 10 years.

In the meantime, it is believed that use of the two-stage system as a producer-gas generator for inplant production of clean fuel gas from coal can be realized earlier. Much will depend upon the incentive provided by current evaluation of the types of end uses described.

New power-generation systems will be developed to help meet the growing requirements of our energy-consuming way of life. Coal, transformed into a clean, non-polluting fuel gas, can continue to help fuel those new systems.

ACKNOWLEDGEMENT

Parts of this paper are based on work carried out at Bituminous Coal Research, Inc., with support from the Office of Coal Research, U.S. Department of the Interior, under Contract No. 14-01-0001-324.

The assistance of Dr. R. L. Zahradnik, Carnegie-Mellon University, and of Dr. E. E. Donath, Consultant, is gratefully acknowledged.

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Pipeline Gas Symposium, New York: p. 67-75, 1966.

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4. Zahradnik, R. L. and R. A. Glenn. The Direct Methanation of Coal. ACS Div. Fuel Chem. Preprints 13 (4): 52-70, 1969.

5. Robson, F. L. and A. H. Giramonti. Advanced-Cycle Power Systems Utilizing Desulfurized Fuels. ACS Div. Fuel Chem. Preprints 14 (2): 79-96, 1970.

6. Curtiss-Wright Corporation. 1000 MW Combined-Cycle Power Generating System. (Curtiss-Wright brochure).

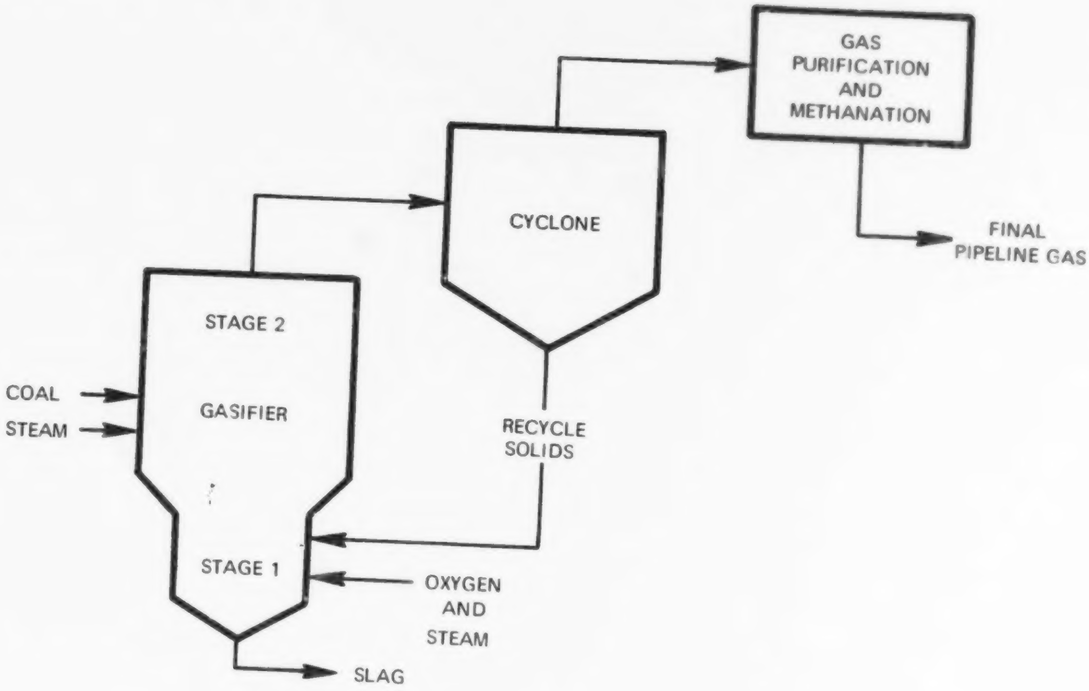

GAS PURIFICATION AND METHANATION
CYCLONE
FINAL PIPELINE GAS
STAGE 2
GASIFIER
COAL
STEAM
RECYCLE SOLIDS
STAGE 1
OXYGEN AND STEAM
SLAG

Figure 1. Simplified flow diagram for two-stage super-pressure gasifier.

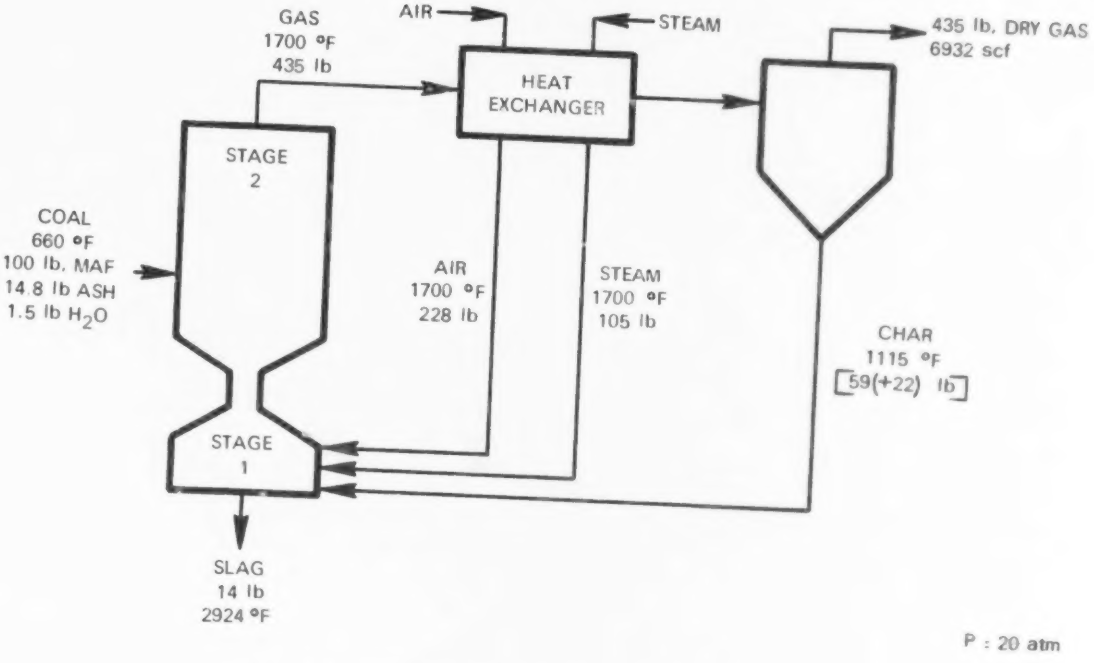
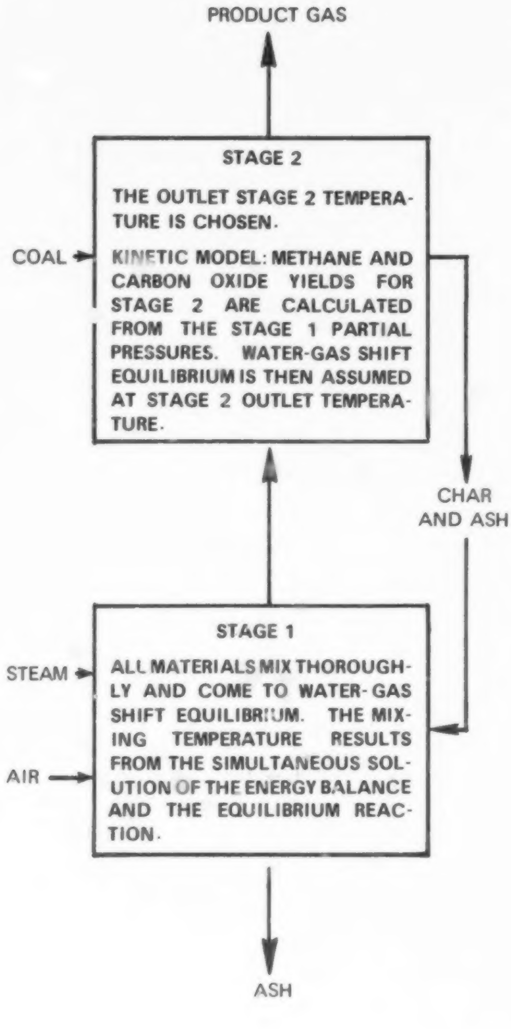

GAS
1700 °F
435 lb
AIR
STEAM
HEAT EXCHANGER
435 lb. DRY GAS
6932 scf
STAGE 2
COAL
660 °F
100 lb. MAF
14.8 lb ASH
1.5 lb H_2O
AIR
1700 °F
228 lb
STEAM
1700 °F
105 lb
CHAR
1115 °F
[59(+22) lb]
STAGE 1
SLAG
14 lb
2924 °F
P : 20 atm

Figure 2. Material balance for air-blown gasifier.



STAGE 2

THE OUTLET STAGE 2 TEMPERATURE IS CHOSEN.

KINETIC MODEL: METHANE AND CARBON OXIDE YIELDS FOR STAGE 2 ARE CALCULATED FROM THE STAGE 1 PARTIAL PRESSURES. WATER-GAS SHIFT EQUILIBRIUM IS THEN ASSUMED AT STAGE 2 OUTLET TEMPERATURE.

STAGE 1

ALL MATERIALS MIX THOROUGHLY AND COME TO WATER-GAS SHIFT EQUILIBRIUM. THE MIXING TEMPERATURE RESULTS FROM THE SIMULTANEOUS SOLUTION OF THE ENERGY BALANCE AND THE EQUILIBRIUM REACTION.

Figure 3. General scheme of mathematical model.

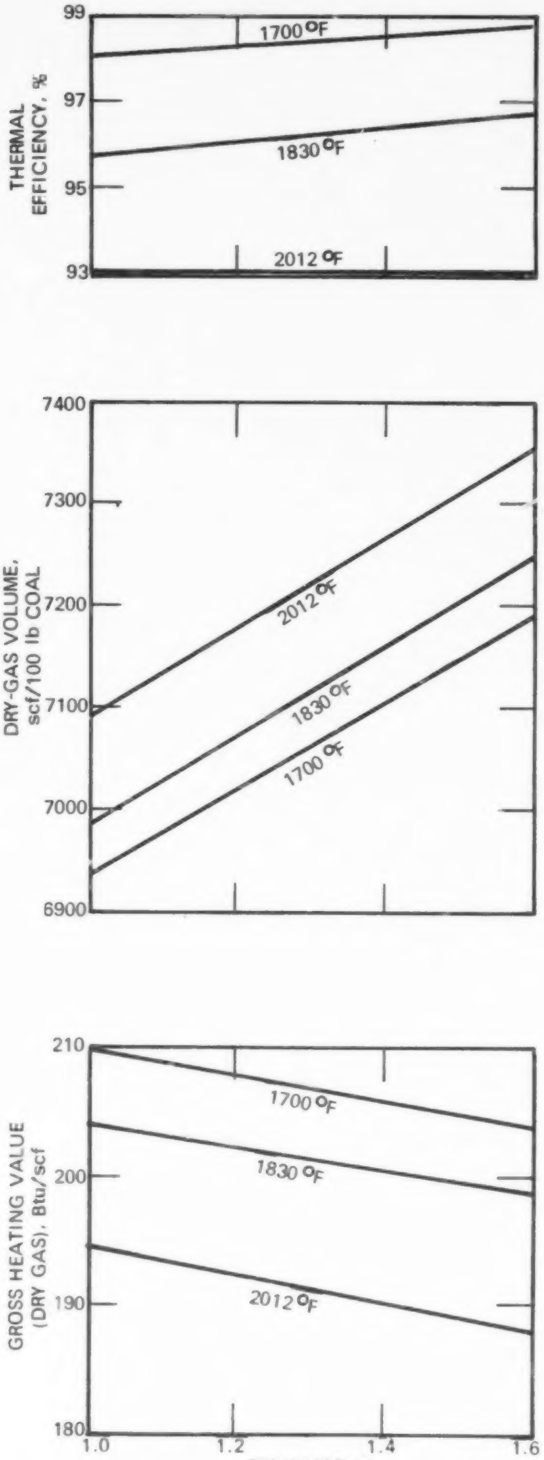


Figure 4. Effect of steam/coal ratio (P = 20 atm).

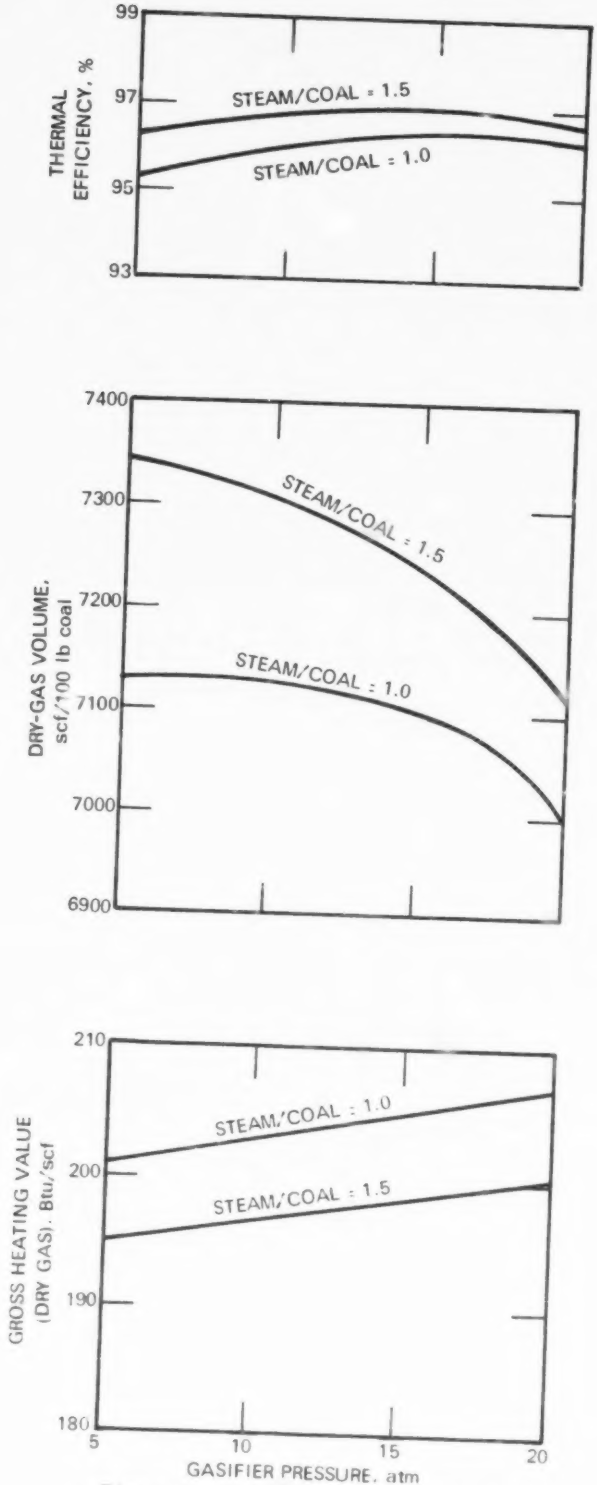


Figure 5. Effect of operating pressure (T = 1830° C).

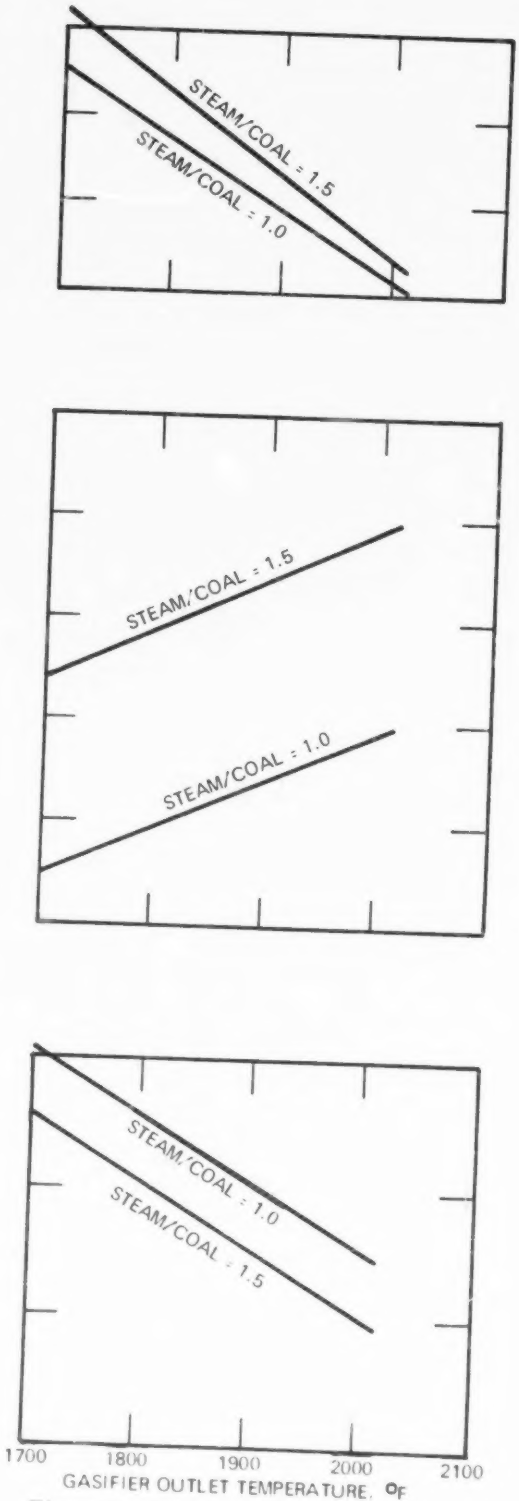


Figure 6. Effect of gasifier outlet temperature (P = 20 atm).

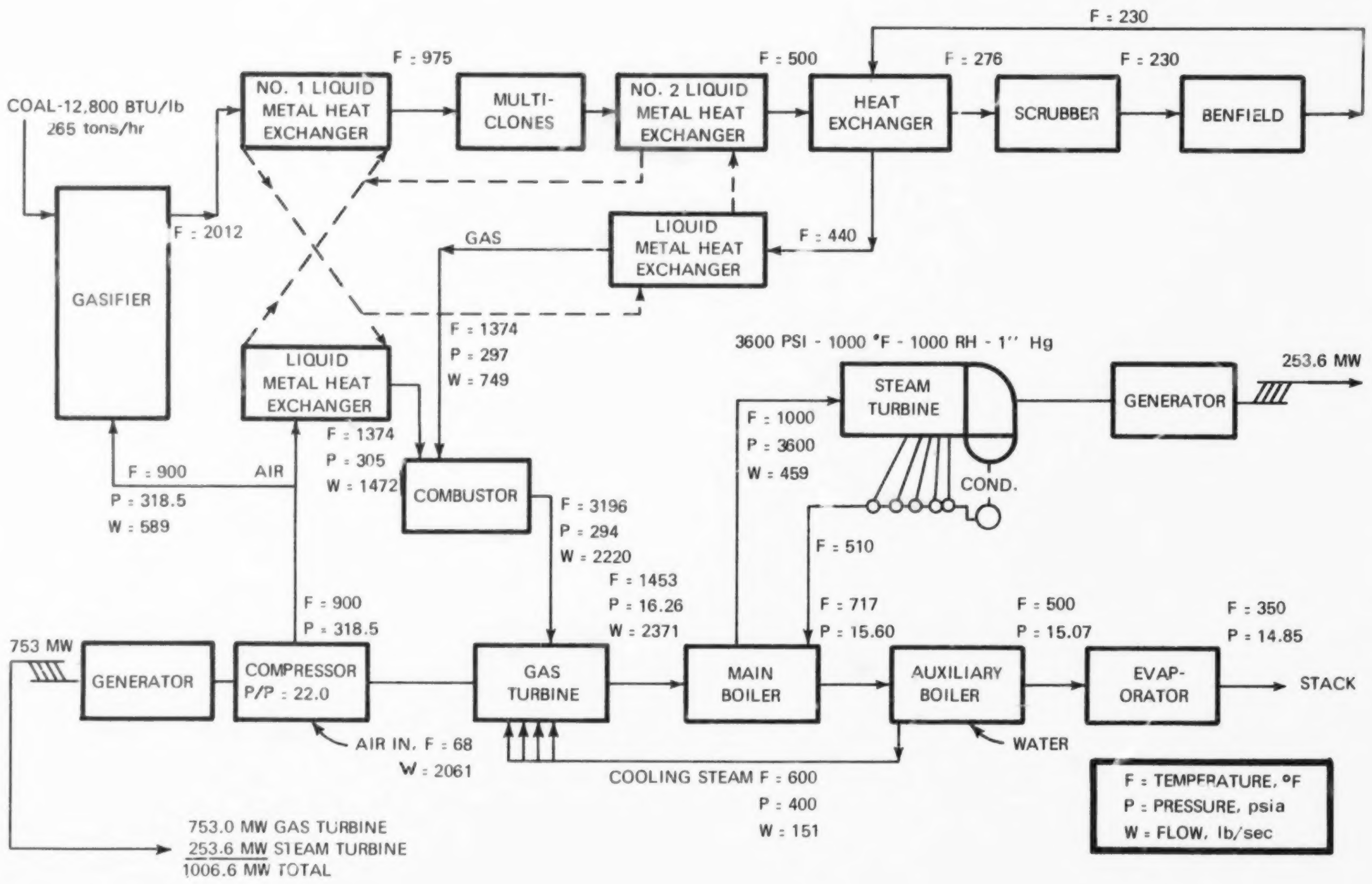

COAL-12,800 BTU/lb
265 tons/hr
GASIFIER
F : 2012
NO. 1 LIQUID METAL HEAT EXCHANGER
F : 975
MULTI-CLONES
NO. 2 LIQUID METAL HEAT EXCHANGER
F : 500
HEAT EXCHANGER
F : 276
SCRUBBER
F : 230
BENFIELD
F : 230
F : 440
LIQUID METAL HEAT EXCHANGER
GAS
F : 1374
P : 297
W : 749
LIQUID METAL HEAT EXCHANGER
F : 1374
P : 305
W : 1472
COMBUSTOR
F : 900
P : 318.5
W : 589
AIR
F : 3196
P : 294
W : 2220
F : 900
P : 318.5
753 MW
GENERATOR
COMPRESSOR
P/P : 22.0
AIR IN. F : 68
W : 2061
GAS TURBINE
F : 1453
P : 16.26
W : 2371
MAIN BOILER
F : 1000
P : 3600
W : 459
3600 PSI - 1000 °F - 1000 RH - 1'' Hg
STEAM TURBINE
COND.
GENERATOR
253.6 MW
F : 510
F : 717
P : 15.60
AUXILIARY BOILER
WATER
F : 500
P : 15.07
EVAP-ORATOR
F : 350
P : 14.85
STACK
COOLING STEAM F : 600
P : 400
W : 151
F : TEMPERATURE, °F
P : PRESSURE, psia
W : FLOW, lb/sec
753.0 MW GAS TURBINE
253.6 MW STEAM TURBINE
1006.6 MW TOTAL

Figure 7. 1000-MW combined-cycle power generating system.

5. REMOVAL OF HYDROGEN SULFIDE FROM SIMULATED PRODUCER GAS AT ELEVATED TEMPERATURES AND PRESSURES

F. G. SHULTZ

U. S. Bureau of Mines

INTRODUCTION

The Morgantown Energy Research Center of the U. S. Bureau of Mines has been investigating the possibility of removing hydrogen sulfide from simulated producer gas near the temperature at which the gas leaves the producer. The temperature range of interest is 1000 - 1500°F. Experimental work has been limited to these temperatures and to pressures between near zero and 100 psig.

Background

We have previously reported[1] that hydrogen sulfide can be removed from this type of hot gas by passing the gas stream through an absorbent bed consisting of sintered pellets. These pellets consist of mixtures of fly ash from a coal-burning power plant at Fort Martin, W. Va., and either technical-grade ferric oxide or "red mud," a residue of the aluminum refining process. These sintered pellets were made by wetting the mixture of the two materials, forming the mixture into nominal 1/4-inch spheres (pellets), drying the pellets, and then firing them in an electric furnace at 1900 - 2300°F for 10-15 minutes. Temperatures required to sinter the materials depended on the composition of the pellets and were higher for a composition containing red mud. Sintering was considered satisfactory when the pellets had been fired enough that the sharp edge of a knife blade would not cut them, but not enough to vitrify the surface.

The absorbent pellets used were previously reported[1] as having H_2S absorption capacities of approximately 12 grams of sulfur per 100 grams of absorbent at 1000°F and 35 weight percent sulfur at 1500°F, when used at space velocities of 2000, bed depths of 15 inches, bed diameters of 1 inch, and absorption pressure near atmospheric. (Space velocity is the number of volumes of gas at standard conditions passing through a unit volume of absorbent per hour.)

Various other metallic oxides, including those of zinc, molybdenum, tin, cerium, lanthanum, and yttrium, were individually sintered from mixtures of the oxide and fly ash. None of these oxides proved to be an effective hydrogen sulfide absorbent when used in this form.

Apparatus and Methods

These absorbents were tested by passing a simulated producer gas through a fixed bed of the absorbent, contained in a stainless-steel pipe, in an electrically heated furnace. Figure 1 shows the basic test system. The test apparatus consisted of compressed-gas cylinders with individual rotameters for blending a gas similar in composition to producer gas; the furnace contains the absorbent and plumbing suitable for passing the gas through the absorbent. Ports allowed for analytical determinations in absorber influent and effluent gas streams. Influent concentrations normally used were 900 - 1000 grains H_2S per 100 cu ft of gas (1.5 percent); effluent concentrations gradually increased from zero to 100 grains per 100 cu. ft., at which concentration the tests were terminated.

DISCUSSION

One unresolved question about the previous work was the effect that tar vapors, which would be present in an actual producer gas, would have on the ability of the abosrbent to remove hydrogen sulfide. A tar-vapor generator was built by partially filling an externally heated 2-inch-diameter cylindrical vessel with a bituminous coal tar made from Pittsburgh-seam coal in the Morgantown Energy Research Center's flash carbonizer. The nitrogen from the flow system was passed over the hot tar to pick up the vapors, then transported through a heated line (900°F) to the bottom of the absorber, where it was mixed with the other producer-gas constituents and then passed through the absorption bed. Gas producers yield about 130,000 cu ft of gas per ton of coal,[2] and an average tar production of about 15 gal. per ton.[3] This is approximately 0.4 ml of liquid tar as vapor per cu ft of gas, or 12,500 ppm by weight. Because nitrogen only was used to sweep the tar-vapor generator, the vapor generator was found to produce 0.8 ml of tar vapor at 600°F. The vapor generator was operated at 600 - 700°F for test work.

Table 1 shows that, at first, the effect of tar vapor seemed to be to increase the capacity of the absorbent. The reason for this increase was unknown at the time. In subsequent tests, Table 2, it was noted that the presence of tar vapors made little difference in absorption capacities until sufficient tar had condensed and been deposited downstream of the absorber to increase both the flow resistance and the pressure in the absorber. Tests made at equal pressures, with and without tar vapor in the gas, indicated equal absorption capacities (tests 110 and 112). These capacities were all determined at gas-flow rates equal to a space velocity of 2000.

Effect of Pressure and Gas Velocity

The effect of increased pressure on absorption was investigated to 10 psig, the limit of the apparatus: Figure 2 illustrates the effect of increasing pressure. Within this pressure range, higher absorption pressures caused an increase in absorption capacity.

Figure 3, relating the effect of increasing space velocity to absorbent capacity, shows that a space velocity of 2000 is near the maximum that would be permissible at 1000°F, whereas at 1500°F removal was still effective at a space velocity of 16,000. These capacities were all determined at 3-psig pressure in the absorption bed.

Table 1. EFFECT OF COAL-TAR VAPORS ON SULFUR ABSORPTION CAPACITIES OF SINTERED JAMAICAN RED MUD FROM REYNOLDS METALS CO.

Test No.	Test temp., °F	Capacity of absorbent, g sulfur/100 g absorbent		Error in sulfur balance,%
		Absorbed from H_2S	Regenerated from SO_2	
99	1000	10.7	12.1	-11.6
100	1250	27.6	24.6	10.9
101	1500	44.1	40.6	7.9
102[a]	1500	56.5	52.3	7.4
103[a]	1250	37.5	34.0	9.3
104[a]	1000	12.0	10.9	9.2

[a] Test gas contained tar vapors.

Table 2. RESULTS OF H_2S ABSORPTION TESTS USING ALCOA MOBILE RED MUD

Test No.	Absorption Temp., °F	Absorption Pressure psig	Weight of sulfur absorbed g sulfur/100g absorbent	Weight of sulfur regenerated g sulfur/100g absorbent	Error in sulfur balance
105[a]	1500	0	6.8	4.1	39.6
106	1500	0	7.4	8.2	9.8
107[a]	1250	0	5.1	4.1	19.6
108	1500	0	7.0	5.6	20.0
109	1500	0	13.3	11.9	10.5
110	1500	3	23.1	22.2	3.9
111	1500	0	14.1	14.8	4.7
112[a]	1500	3	23.6	23.8	0.8
113	1500	6	29.1	27.9	4.1
114	1500	10	32.8	28.5	13.1

[a] Test gas contained tar vapors.

Sintering Conditions

The effect of sintering temperatures on capacity was thought worthy of investigation. It was found, however, that these absorbents have a fairly narrow temperature range in which sintering will occur within a reasonable time period. Hurricane Creek (H. C.) Jamaican red mud will sinter in 2 hours at 2200°F, and in 10 minutes at 2300°F, but it quickly develops a vitreous surface at 2400°F. Materials incorporated into a fly ash mixture generally sinter best at 1700 - 2000°F, but again a temperature range of only about 200°F can cause the difference between excessively long sintering times and the rapid formation of a glassy surface for any specific composition.

Durability of Absorbents

All previous work had been done using a 1-inch-diameter stainless steel pipe, type 304 or 316, as an absorption tube. This unit was too small to permit easy unloading of absorbent pellets. The inside walls of the pipe react with the gases used in this work, causing a scale formation that gradually (but partially) envelops the pellets in contact with the pipe wall. After several tests the pellets could be removed only by rodding out the pipe and crushing the pellets.

To assess the durability of absorbents better, a split-shell furnace with a bore sufficient to hold a 2.5-inch-diameter pipe was substituted for the original furnace. In the 1-inch-diameter unit, some absorbents disintegrated into dust, some fused into a mass, and others appeared fairly durable. The more durable absorbents tested in the 1-inch unit were: (1) sintered Fort Martin fly ash, (2) a sintered mixture of 25 percent Fe_20_3 and 75 percent Fort Martin fly ash, and (3) a sintered mixture of 40 percent H.C. Jamaican red mud and 60 percent Fort Martin fly ash. Although the fly ash alone demonstrated good durability in the smaller unit, no additional work was done with it because it had relatively low sulfur-absorption capacity. Both the other absorbents were used in the larger diameter absorber; however, each was tested through six H_2S absorption-air regeneration cycles.

Because the larger unit permitted easy unloading, weights of absorbents before and after testing could be determined. After testing, the red-mud-containing absorbent had lost 3 percent of its weight, probably as dust; 10 percent of the pellets had fused into two separate lumps; and a large amount of pellet fracture had occurred. The Fe_20_e-containing absorbent did not lose weight, no fusion between pellets had resulted, and only a small amount of pellet fracture was observed. Absorption capacity was somewhat lower in the larger diameter unit: only 16 weight percent at a space velocity of 500, a temperature of 1500°F, and a 10-inch bed depth. The reason for this lower absorption capacity is being investigated.

Absorption at Higher Pressures

The effect of higher pressures (up to 100 psig) was investigated in the 2.5-inch-diameter unit with a 5-inch bed depth of the sintered 25 percent Fe_2O_3-75 percent fly-ash absorbent that had been used in ten previous tests. (Table 3 shows the results.) Premixed gas in pressurized cylinders was used for the 100-psig test. Absorption tests were made at 1500°F and pressures of 15, 30, 45, 50, and 100 psig. Space velocities used ranged from 1100 to 4400. Capacities varied from 15.3 to 16.3 weight percent, except for a 45-psig test in which flow was erratic. This capacity was a 20 percent increase above that obtained at 1500°F and 3 psig with a 5-inch bed depth, but no trend of capacity change as a function of pressure was indicated in this region. The effect of pressure on absorption capacity for this system becomes asymptotic at 10 to 15 psig, within the region of space velocity utilized.

The 25-percent Fe_2O_3 and fly-ash absorbent has been tested through 15 cycles of absorption and regeneration (Table 3): the only evidence of degradation has been some fracturing of pellets and the fusion of 2 percent of the pellet weight into one lump during the series of absorptions at elevated pressure. The maximum temperature at which this absorbent is durable is 1500°F; at 1600°F, fusion between pellets becomes severe.

Table 3. H_2S ABSORPTION CAPACITIES OBTAINED DURING 15 ABSORPTION-REGENERATION CYCLES WITH A 25 PERCENT Fe_2O_3-75 PERCENT FLY-ASH SINTERED ABSORBENT[a]

Absorption temp., °F	Gas flow, scfh	H_2S g/100 ft^3	Absorption pressure, psig	Space velocity	Duration of test, hr	Sulfur absorbed, wt %
1000	15	1030	3	2000	1.5	6.2
1500	15	890	3	2000	3.5	12.0
1250	15	900	3	2000	2.5	9.0
1500	15	960	3	2000	3.5	13.5
1250	15	910	3	2000	3.0	11.0
1000	15	945	3	2000	2.25	8.4
1000	15	990	3	1000	3.25	6.1
1250	15	1080	3	1000	4.33	8.9
1500	15	1040	3	1000	6.5	12.8
1000	15	1010	3	1000	4.75	9.1
1500	30	1100	15	2000	4.0	16.3
1500	45	1050	30	3000	2.5	15.3
1500	40	1050	45	2670	2.25	12.2
1500	66	1050	50	4400	1.75	15.7
1500	16.5	393	1000	1100	16.75	15.8

[a] First 6 tests made with 2.5-inch bed depth; last 9 tests, with 5-inch.

Regeneration of Absorbent

Regeneration of these absorbents with air has been found to proceed best when initiated at about 1200°F. Sulfur dioxide is liberated in concentrations of 5-8 volume percent until about 60-70 percent of the absorbed sulfur is burned off at air-input space velocities of 2000. The concentration of sulfur dioxide then rapidly declines but never becomes zero, even after a regeneration time of 80 hours. An SO_2 concentration of about 50 ppm is reached after 24 hours of regeneration, and the absorbent is then reused for H_2S absorption. During regeneration a small quantity of elemental sulfur is also liberated.

Water vapor is formed during the absorption step and condenses in lines downstream of the absorber. No doubt some SO_3 forms during regeneration and combines with this condensed water, because sulfuric acid has been identified in the effluent system.

CONCLUSIONS

Conclusions deduced from work to date follow:

1. With the type of sintered absorbent described here, H_2S can be absorbed from a producer gas at near the exit temperature of the gas, thus preserving the sensible heat of the gas for producing work.
2. Absorbents containing more than 37 percent Fe_2O_3 degrade more rapidly than those with lesser amounts of Fe_2O_3.
3. Degradation consists of either pellet fusion or disintegration into dust.
4. The maximum temperature at which this absorbent can be used is 1500°F.
5. Tar vapors in the producer gas do not adversely affect the capacity of the absorbent for H_2S during the absorption time intervals used in this work.
6. Absorptive capacity increases with pressures up to 15 psig.

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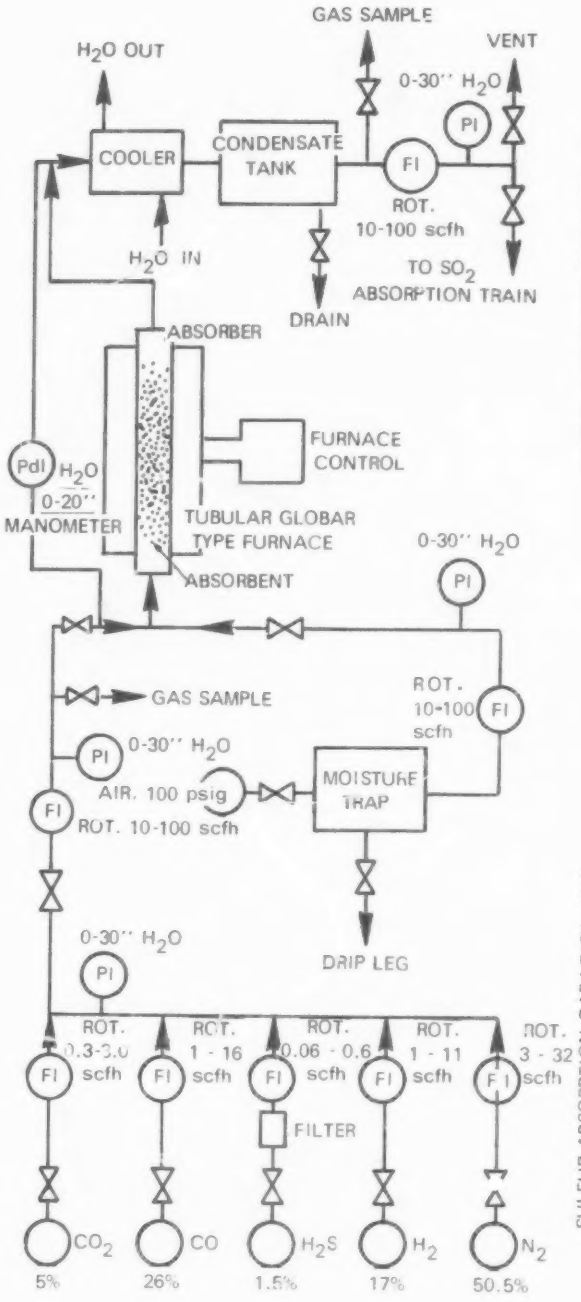
GAS SAMPLE
H_2O OUT
VENT
0-30'' H_2O
PI
COOLER
CONDENSATE TANK
FI
ROT. 10-100 scfh
H_2O IN
TO SO_2 ABSORPTION TRAIN
DRAIN
ABSORBER
FURNACE CONTROL
PdI
H_2O 0-20''
MANOMETER
TUBULAR GLOBAR TYPE FURNACE
ABSORBENT
0-30'' H_2O
PI
ROT. 10-100 scfh
FI
GAS SAMPLE
PI
0-30'' H_2O
AIR, 100 psig
MOISTURE TRAP
FI
ROT. 10-100 scfh
DRIP LEG
0-30'' H_2O
PI
ROT. 0.3-3.0 scfh
ROT. 1 - 16 scfh
ROT. 0.06 - 0.6 scfh
ROT. 1 - 11 scfh
ROT. 3 - 32 scfh
FI
FILTER
CO_2
5%
CO
26%
H_2S
1.5%
H_2
17%
N_2
50.5%

Figure 1. Flowsheet for removal of sulfur from hot producer gas.

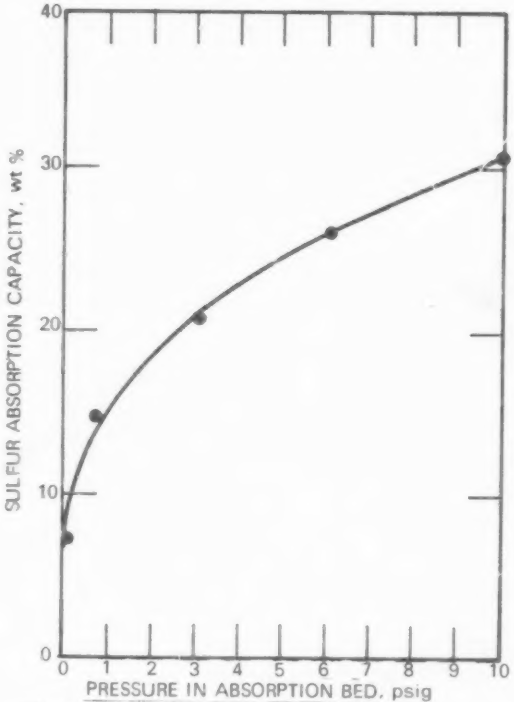

Figure 2. Sulfur absorption capacity as a function of absorption-bed pressure for Alcoa mobile red mud.

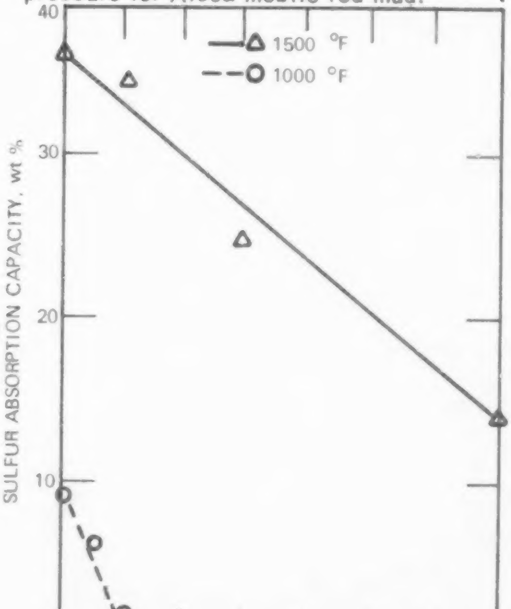

Figure 3. Sulfur absorption capacity as a function of space velocity for Reynolds H. C. Jamaican red mud.

6. GASIFICATION OF SOLID PARTICLES CONTAINING CARBON

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INTRODUCTION

Fluidized-bed gasifiers have been shown to possess technical and economical advantages over conventional coal-gasifying devices in solving air pollution problems resulting from the use of high-sulfur coal. However, the complex phenomena of chemical reaction, heat and mass transfer on solid particles, and the interaction of gas and solid flow patterns within the fluidized bed have hampered design development. Researchers in recent years have, however, provided insight into the rate of processes occurring on a single particle and with the phenomena associating bubbles in fluidized beds. It is the purpose of this paper to present the method by which realistic mathematical model describing a single-particle phenomenon can be formulated and how such information can be incorporated into a fluidized-bed model that accounts for the bubble growth and coalescence in order to arrive at a more reliable design for fluidized-bed gasifiers.

SINGLE-PARTICLE SOLID-GAS REACTION MODELS

The reaction models for single-particle solid-gas reaction systems can be generally divided into three categories: (1) unreacted-core-shrinking model, (2) homogeneous model, and (3) zone-reaction model.

If the porosity of the unreacted solid is very small and the solid is practically impervious to gaseous reactants, the reaction will occur at the interface between the unreacted solid and the porous-product layer. Under such conditions the unreacted-core-shrinking model is applicable. Figure 1 shows the solid and gaseous reactant concentration profiles for this model. This model is also applicable when the chemical reaction rate is very rapid in comparison with the diffusion rate of the gaseous reactant. The zone of reaction in such a case is narrowly confined to the interface between the unreacted solid and the product solid.

On the other hand, if the solid is porous enough for the gaseous reactant to diffuse freely into the interior of the solid, the unreacted-core-shrinking model is no longer applicable. In such a case the reaction may be said to occur, in a macroscopic sense, homogeneously throughout the solid to produce a gradual and uniform variation in solid-reactant concentration in the particle. The homogeneous model describes this situation closely.

In most actual cases, however, the solid-gas reactions cannot be completely represented by either of the first two models mentioned above, which represent two extreme cases in the solid-gas reaction systems. The majority of the solid-gas reactions probably fall between these two extremes.

The zone-reaction model[1,2] is a more general model for solid-gas reactions because it displays an intermediate between the unreacted-core-shrinking and homogeneous models, as shown schematically in Figure 2. In fact, the unreacted-core-shrinking and homogeneous models can be derived as special cases from the zone-reaction model.[3]

In the zone-reaction model, the kinetic behaviors depend on the solid structures. Figure 2 also shows three types of solid structures:

1. Volumetric reaction model: the solid particle is composed of small grains of solid reactant embedded in inert material.
2. Grain model: the solid particle is composed of small grains, each of which reacts according to the unreacted-core-shrinking model.
3. Pore model: the reaction occurs at the surface of the pores.

The effects of temperature on the reaction rate and on the concentration profiles of gaseous reactant (A) and solid reactant (S) are summarized schematically in Figure 3. In the lowest temperature region, V, of the zone-reaction model, the concentration distribution of both gaseous and solid reactants are uniform—a limiting case in which the homogeneous model is applicable. The reaction rate in this temperature region is controlled by that of individual grains or pores. In the highest temperature region, I, on the other hand, the gaseous reactant concentration at the boundary between the reaction zone and the product layer is practically zero, so that the reaction rate is controlled by the diffusion of gas through the product layer.

Note that in temperature regions I and III of the zone-reaction model, the gaseous reactant concentration profiles in the product layer are similar to those for the unreacted-core-shrinking model as shown in Figure 3(b).

Because the unreacted-core-shrinking model has wide application for many investigators, and because the mathematical treatment is relatively simple in comparison with the zone-reaction model, a brief discussion of the unreacted-core-shrinking model is given below, with emphasis on the effects of heat and mass transfer on reaction rate in terms of the effectiveness factor. The detailed mathematical treatments can be found elsewhere.[3,4,5]

The effectiveness factor is defined as follows:

$$\eta_s = \frac{\text{actual (overall) reaction rate}}{\text{reaction rate obtainable when the reaction site is exposed to the gas concentration and temperature of the bulk-gas phase}}$$

Because the denominator is a constant, the effectiveness factor is, in effect, a dimensionless surface-reaction rate.

Figures 4 and 5 show plots of effectiveness factor versus fractional solid reactant conversion, and compare unsteady-state heat transfer solution to that of pseudo-steady-state. Figure 4 indicates a case in which the pseudo-steady-state analysis could lead to an erroneous conclusion. The chemical-reaction-controlling region would never have been realized if the pseudo-steady-state analysis had been used. The unsteady-state analysis, on the other hand, shows that chemical reaction could be rate-controlling when the initial temperature of the particle is sufficiently low. Figure 5 depicts the effects of the heat capacity of the unreacted core and the heat of reaction on thermal instability. The occurrence of thermal instability is less likely when the heat capacity is high.

Figure 6 illustrates experimental results that were computed on the basis of the unreacted-core-shrinking model. The corresponding numerical solutions (solid, dotted, and dashed lines) are also shown in the figure for comparison. The experiment was performed in a thermabalance (shown in Figure 7) by burning a single solid sphere in a stream of heated air. The solid pellet was prepared by mixing the desired proportion of activated charcoal with aluminum oxide, which serves as an inert porous medium. Sodium silicate (waterglass), diluted with the proper amount of water, was used as a binder in forming the pellets. The particle was heated at 700°C in an inert atmosphere prior to the combustion test to remove moisture and to ensure no weight loss from inert solid during the test.

"Extinction" occurred at about 85 percent of solid conversion. Note that the unsteady-state heat transfer analysis (solid line) describes more closely the experimental result than the pseudo-steady-state analysis (dashed line) at the initial and transition stages. The values of the parameters used in the correlation are independently estimated from empirical correlations.

Figure 8 is an example of a triangular diagram that shows the reaction path of an endothermic reaction. The corresponding effectiveness factor versus conversion plot is shown in the Figure 8 inset. The main advantage of a triangular diagram is that it shows the magnitude of each resistance during the history of a reaction.

In the following discussion we consider only the case in which solid particles react with fluidizing gas while maintaining their original size because of the formation of inert solid product. The gasification of coal, the roasting of sulphide ores, and the reduction of iron ores are examples of this situation. The following stoichiometric equation can be used to represent these reactions.

$$aA(\text{gas}) + S(\text{solid}) \rightarrow \text{gaseous and/or solid products} \quad (1)$$

The proposed calculation method[6] assumes that solids follow the shrinking-core model and that the overall conversion rate is controlled by a chemical reaction step; in the unreacted-core-shrinking model, the reaction is confined at the surface of the core, which recedes from the outer surface toward the interior of the particle. When diffusion through the product layer becomes rate-controlling, or when other single-particle reaction models are used, the conversion-versus-time expressions must be changed accordingly for our calculations.

The reaction of a gaseous component by a first-order irreversible reaction can be given as:[6]

$$-\frac{1}{4\pi r_c^2 a} \bullet \frac{dN_A}{dt} = -\frac{1}{4\pi r_c^2} \bullet \frac{dN_s}{dt} = k_c C_A \quad (2)$$

where r_c is the radius of unreacted core and k_c is the rate constant for the reaction.

When the reaction is carried out in the bulk-phase reactant-gas concentration, C_A, the extent of conversion, X_S, of a particle having radius R is given by:

$$\frac{t}{\tau} = 1 - \frac{r_c}{R} = 1 - (1 - X_s)^{1/3} \quad (3)$$

where time for complete conversion, τ, is:

$$\tau = \frac{\rho_p d_p}{a k_c C_{Ao}} \quad (4)$$

When the resistances of the chemical reaction step and of diffusion through the ash layer are comparable, the rate constant, k_c, is replaced by $\overline{k}$, defined by:

$$\frac{1}{\overline{k}} = \frac{1}{k_c} + \frac{d_p}{12 D_{eA}} \quad (5)$$

Next, let us consider a reactor with a constant feed rate of both solids and gas, the solids being of uniform size and complete mixing. Because the conversion of an individual particle of solid depends on its length of stay in the reactor, the mean conversion, $\overline{X}_s$, of the exit stream of solids is given by:

$$1 - \overline{X}_s = \int_{t=0}^{\tau} (1 - X_s)\, E(t)\, dt \quad (6)$$

where the exit age distribution function for a reactor of complete mixing is:

$$E(t) = \frac{1}{\overline{t}}\, e^{-t/\overline{t}} \quad (7)$$

When chemical reaction is the rate-controlling step in a shrinking-core particle, substitution of equations (3) and (7) into equation (6) and subsequent integration yield:

$$1 - \overline{X}_s = 1 - 3\left(\frac{\overline{t}}{\tau}\right) + 6\left(\frac{\overline{t}}{\tau}\right)^2 - 6\left(\frac{\overline{t}}{\tau}\right)^3 \left[1 - \exp\left(-\tau/\overline{t}\right)\right] \quad (8)$$

Gas and Solid Flows Based on the Bubble Assemblage Model

Let us summarize the essentials of the bubble assemblage model[7] as follows:

A fluidized bed may be approximately represented by "N" numbers of compartments in series. The height of each compartment is equal to the size of each bubble at the corresponding bed height. Each compartment is considered to consist of the bubble phase and the emulsion phase.

The bubble phase is assumed to consist of spherical clouds; the cloud diameter can be derived from:

$$\left(\frac{d_c}{d_b}\right)^3 = \frac{u_{br} + 2u_{mf}/\epsilon_{mf}}{u_{br} - u_{mf}/\epsilon_{mf}} \qquad (9)$$

The size of the bubble diameter along the bed height is approximated by:

$$d_b = 1.4\,\rho_p d_p \left(\frac{u_o}{u_{mf}}\right) h + d_o \qquad (10)$$

where d_o is the bubble diameter just above the distributor. The rising velocity of a bubble is given by:

$$u_b = u_o - u_{mf} + 0.711(gd_b)^{1/2} \qquad (11)$$

From an arithmetic average of bubble sizes, the height of the i-th compartment can be expressed as:

$$\Delta hi = 2d_o \frac{(2+m)^{i-1}}{(2-m)^1} \qquad (12)$$

where $m = 1.4\,\rho_p d_p (u_o/u_{inf})$.

The voidage distribution assumes that up to L_{mf}, the voidage, ϵ, is uniform, while above L_{mf}, ϵ increases linearly along the bed height. Then the number of bubbles, n, in the i-th compartment is given by:

$$n = \frac{6St}{\pi(\Delta h_i)^2} \cdot \frac{\epsilon - \epsilon mf}{1 - \epsilon mf} \qquad (13)$$

Therefore, the volume of bubbles, clouds, and emulsion in the i-th compartment can be calculated, respectively:

$$V_{bi} = n \cdot \frac{\pi}{6}(\Delta h_i)^3 \qquad (14)$$

$$V_{ci} = n \cdot \frac{\pi}{6}(\Delta h_i)^3 \left(\frac{3u\, mf/\epsilon mf}{u_{bri} - u_{mf}/\epsilon mf}\right) \qquad (15)$$

$$V_{ei} = S_t \Delta h_i - V_{bi} - V_{ci} \qquad (16)$$

The overall interchange coefficient of the gas between the bubble and the emulsion phase based on a unit volume of gas bubbles may be given by the following experimental relation:

$$(K_{be})_b = 11/d_b \qquad (17)$$

The solid interchange coefficient between both phases is assumed to be given by:

$$(K_{be})_{bs} = 3\,\frac{(1-\epsilon mf\; u_{mf}\, u_b}{\epsilon mf \cdot u_{br} \cdot d_b} \qquad (18)$$

In order to describe the distribution of solids, let us define, for convenience, the following quantities:

$$\gamma_c = \frac{\text{volume of solids dispersed in clouds and wake}}{\text{volume of bubbles}}$$

$$\gamma_e = \frac{\text{volume of solids in emulsion}}{\text{volume of bubbles}}$$

The upward motion of the solids, as a part of the wake of the bubbles rising from the i-th compartment to the (i+1)-th compartment, sets up a circulation in the bed with downward movement of solids in the emulsion phase from the (i+1)-th compartment to the i-th compartment. When the solids are fed to the bottom of the bed at a constant volumetric flow rate, W, the total upward flow rate, W_b, from the i-th compartment is given by:

$$W_{bi} = \left(\frac{W}{S_t} + \frac{\alpha U_{bi} S_{bi}}{S_{bi}}\right) S_{bi} \qquad (19)$$

where $S_{bi} = \eta \cdot \frac{\pi}{4}(\Delta h_i)^2$

The total downward flow rate, W_e, from the (i+1)-th compartment to the i-th compartment is given by:

$$W_{e(i+1)} = \left(\frac{\alpha \cdot U_{bi} S_{bi}}{S_t - S_{bi}} - \frac{W}{S_t}\right)\left(S_t - S_{bi}\right) \qquad (20)$$

If the solids are fed at the top of the bed and withdrawn from the bottom, W must be replaced by -W in the above equations. These relations mentioned above are shown schematically in Figures 9 and 10.

NOMENCLATURE

The following list of terms defines expressions used throughout this paper, in both the text and the figures.

Term	Definition
a	Stoichiometric coefficient
A	$C_{Ao} D_{eA(To)} C_{pe} / a C_{So} k_e$, ratio of mass to thermal diffusivities in ash layer
C_A	Concentration of gaseous reactant A in ash layer, C_{Ac}
C_{Ac}	Concentration of gaseous reactant A to unreacted-core surface
C_{Am}	Concentration of gaseous reactant A at boundary between reaction and diffusion zones
C_{Ao}	Concentration of gaseous reactant A in bulk gas phase
C_{As}	Concentration of gaseous reactant A at outer surface of particle, mole/L^3
C_{gbi}	Gas-phase concentration in the bubble in the i-th compartment
C_{gei}	Gas-phase concentration in the emulsion in the i-th compartment
C_{pc}	Heat capacity of unreacted core, H/MT
C_{pe}	Volumetric heat capacity of ash layer, H/L^3T
C_S	Concentration of solid reactant S
C_{So}	Initial concentration of solid reactant S
C_{Ss}	Concentration of solid reactant S at outer surface of particle, mole/L^3
d_b	Bubble diameter, cm
d_{bi}	d_b in the i-th compartment, cm
d_c	Diameter of cloud, cm
d_o	Bubble diameter just above the distributor, cm
d_p	Particle diameter, cm
D_{eA}	Effective diffusivity of component A in ash layer, L^2/θ
E	Activation energy of reaction rate constant
E_a	Apparent activation energy, H/mole
E_D	Activation energy in temperature range where diffusion is controlling
E_{ks}	$1/2\ E_{kv}$
E_{kv}	Activation energy in temperature range where reaction rate is controlling
g	Acceleration of gravity, cm/s^2
G	$\rho_c C_{pc} T_o / a C_{So}(-\Delta H)$, ratio of enthalpy of unreacted core to heat of reaction
h	Distance from the distributor, cm
h_C	Convective heat transfer coefficient, $H/L^2\theta T$
Δh_i	Height of the i-th compartment, cm
h_R	Radiational heat transfer coefficient, $H/L^2\theta T^4$
H	$\dfrac{U_c}{\exp\left[\frac{E}{\mathcal{R}T_o}\left(1 - \frac{1}{U_c}\right)\right]} - 1$
ΔH	Heat of reaction per mole of gaseous reactant A, H/mole
$\overline{k}$	Overall rate constant, cm/s
k_e	Effective thermal conductivity of ash layer, $H/L\theta T$
k_s	Rate constant for surface reaction defined in equation (2), cm/s
$(K_{be})_b$	Interchange coefficient of gas between bubble and emulsion, s^{-1}
$(K_{be})_{bi}$	$(K_{be})_b$ in the i-th compartment
$(K_{be})_{bs}$	Interchange coefficient of solid between bubble and emulsion based on volume of a bubble, s^{-1}
k_{mA}	Mass transfer coefficient of component A across gas film, L/θ
L_f	Height of fluidized bed, cm
L_{mf}	Bed height at minimum fluidizing conditions, cm
m	Order of reaction for solid reactant

M	$\phi_s \frac{r_c^2}{R^2} \left(\frac{1}{N_{Sh}} + \frac{R}{r_c} - 1\right)$
n	Number of bubbles in i-th compartment; also order of reaction for gaseous reactant
N_A, N_B	Gramme moles of A and B
$(N_{Nu})_C$	Rh_C/k_e, modified Nusselt number of convective heat transfer
$(N_{Nu})_R$	$Rh_R T_o^3/k_e$, modified Nusselt number for radiational heat transfer
N_{Re}	Reynolds number
N_{Sh}	$Rk_{mA(T_o)}/D_{eA(T_o)}$, modified Sherwood number
r	Distance from the center of sphere (or from solid surface) to reaction surface
r_c	Radius of unreacted core, L
r_m	r at boundary between reaction zone and diffusion zone, L
R	Radius of a particle, cm
$\mathcal{R}$	Gas constant, H/mole T
S_{bi}	Cross sectional area of bubble phase in the i-th compartment, cm^2
S_t	Cross sectional area of the bed, cm^2
$\bar{t}$	Mean residence time of particles, s
T	Temperature
T_c	Temperature at unreacted-core surface
T_i	Initial particle temperature
T_o	Temperature at bulk gas phase
T_s	Temperature at outer surface of particle
u_b	Velocity of rising bubble, cm/s
u_{bi}	Velocity of rising bubble in the i-th compartment, cm/s
u_{br}	Velocity of bubble with respect to emulsion ahead of it, cm/s
u_e	Velocity of emulsion phase, cm/s
u_{mf}	Minimum fluidizing velocity, cm/s
u_o	Superficial gas velocity, cm/s
U_c	T_c/T_o
V_{bi}	Volume of bubble phase in the i-th compartment, cm^3
V_{ci}	Volume of cloud region in the i-th compartment, cm^3
V_{ei}	Volume of emulsion phase in the i-th compartment, cm^3
W	Volumetric feed and outflow rate of solids, cm^3
W_{bi}	Volumetric upward flow rate from the i-th compartment, cm^3/s
W_{ei}	Volumetric downward flow rate from the i-th compartment, cm^3/s
X	Fractional conversion of solid reactant S
$\overline{X}_s$	Mean fractional conversion of solid
β	$C_{Ao} D_{eA(T_o)} (-\Delta H) \mathcal{R}/k_e E$
ϵ	Void fraction in a bed as a whole
ϵ_{mf}	Void fraction in a bed at minimum fluidization
η_s	Effectiveness factor based on surface reaction
η_v	Effectiveness factor based on volume reaction
ρ_c	Density of unreacted core, M/L^3
ρ_p	Density of solid, g/cm^3
ϕ_s	$aRk_{s(To)} C_{Ao}^{n-1} C_{So}^{m} / D_e A(T_o)$, modified Thiele modulus
τ	Time for complete conversion of a single particle, s

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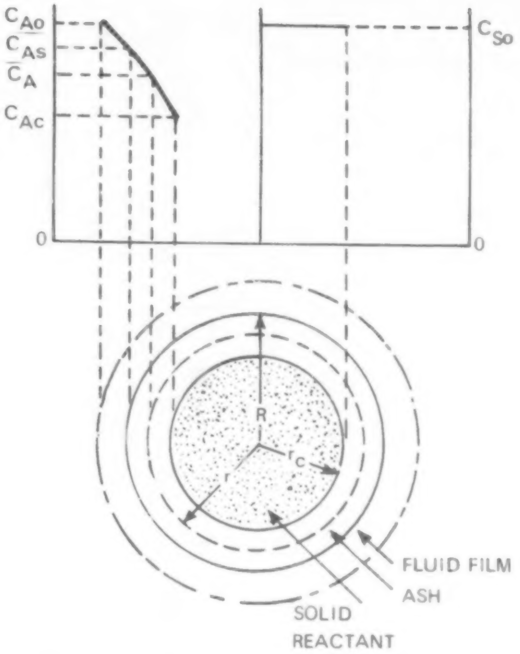

C_{Ao}
$\overline{C_{As}}$
$\overline{C_A}$
C_{Ac}
0
C_{So}
0
R
r_c
r
FLUID FILM
ASH
SOLID REACTANT

Figure 1. Concentration profile for the unreacted-core-shrinking model.[3]

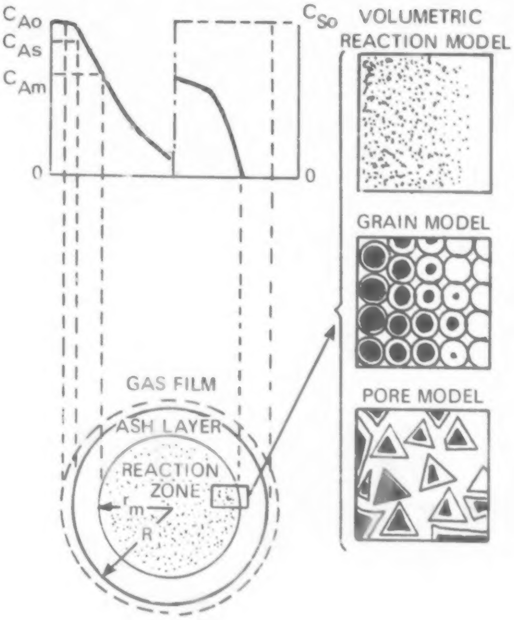
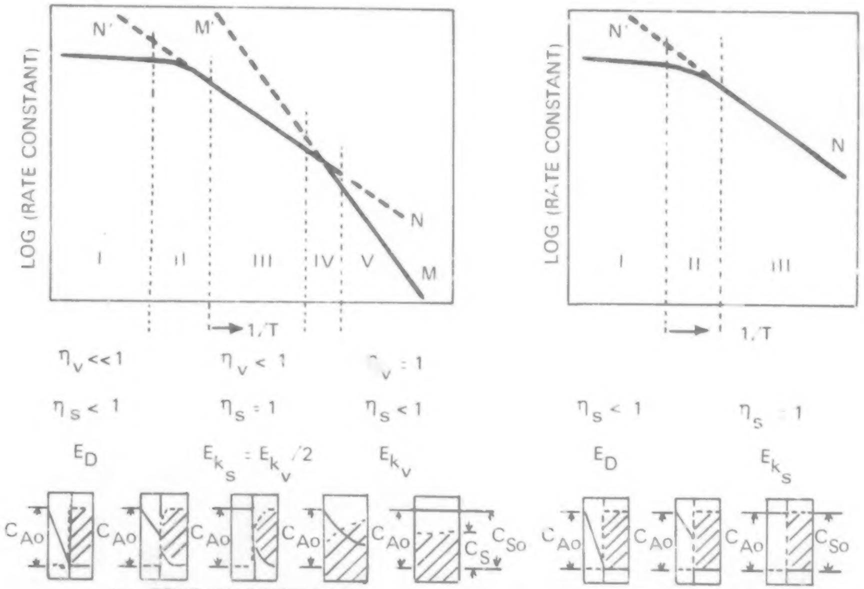

C_{Ao}
C_{As}
C_{Am}
0
C_{So}
0
VOLUMETRIC REACTION MODEL
GRAIN MODEL
PORE MODEL
GAS FILM
ASH LAYER
REACTION ZONE
r_m
R

Figure 2. Concentration profile and solid structure in the particle.[1]



(a) ZONE-REACTION MODEL (b) UNREACTED-CORE MODEL

Figure 3. Schematic diagram representing characteristic behavior of solid-gas reaction systems under various temperature regions.[1]

$\beta = 0.005$

$N_{Sh} = 100$

$\frac{E}{R\,T_o} = 25$

$\Phi_s = 0.139$

$(N_{Nu})_C = 1$

$n = 1$

$A = 0$

$(N_{Nu})_R = 0$

$G = 0.5$

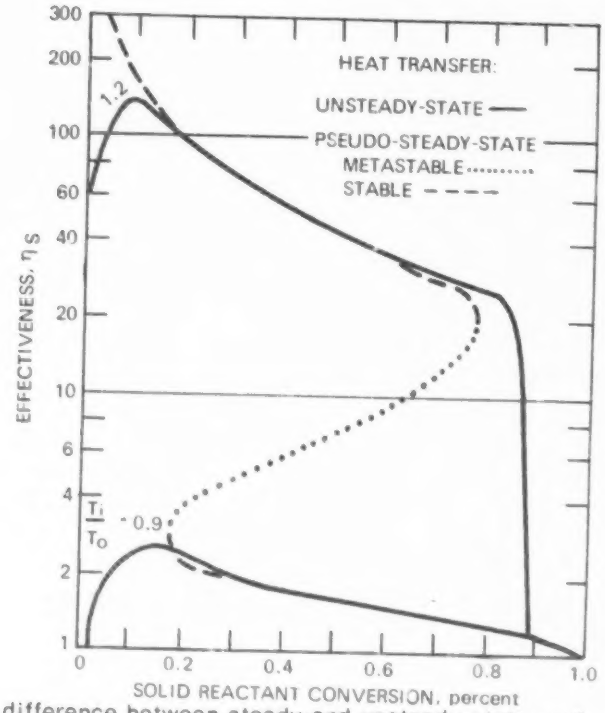


Figure 4. An example of the difference between steady-and unsteady-state analysis.[4]

$\beta = 0.02$

$N_{Sh} = 100$

$\frac{E}{R\,T_o} = 25$

$\Phi_s = 0.139$

$(N_{Nu})_C = 10$

$n = 1$

$A = 0$

$(N_{Nu})_R = 0$

$\frac{T_i}{T_o} = 0.95$

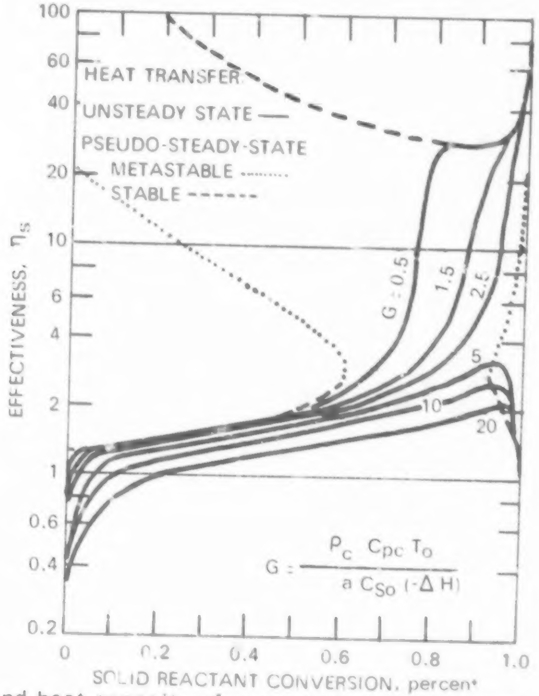
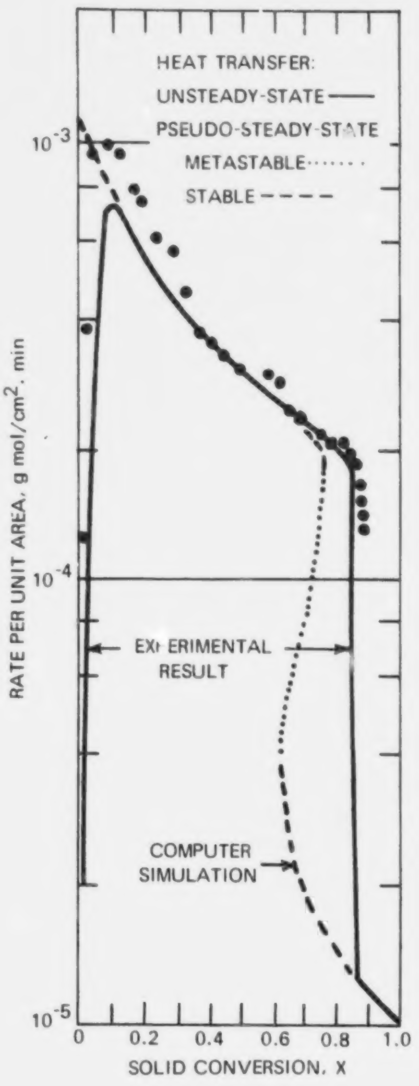


Figure 5. Effect of heat of reaction and heat capacity of unreacted core on thermal instability in terms of G.[4]



T_o = 442 °C

T_w = 573 °C

T_i = 27 °C

N_{Re} = 393

R = 1.244 cm

C_{So} = 0.0225 g mol/cm^3

ε = 0.556

D_{eA} = 0.321 cm^2/sec

K_e = 0.0032 cal/sec-cm-°C

E = 35,700 cal/g mol

$K_{s\,(To)}$ = 5.16×10^{-4} cm/sec

Figure 6. Comparison of experimental result and computer simulation for oxidation of carbon in porous solid, using unreacted-core-shrinking model.4

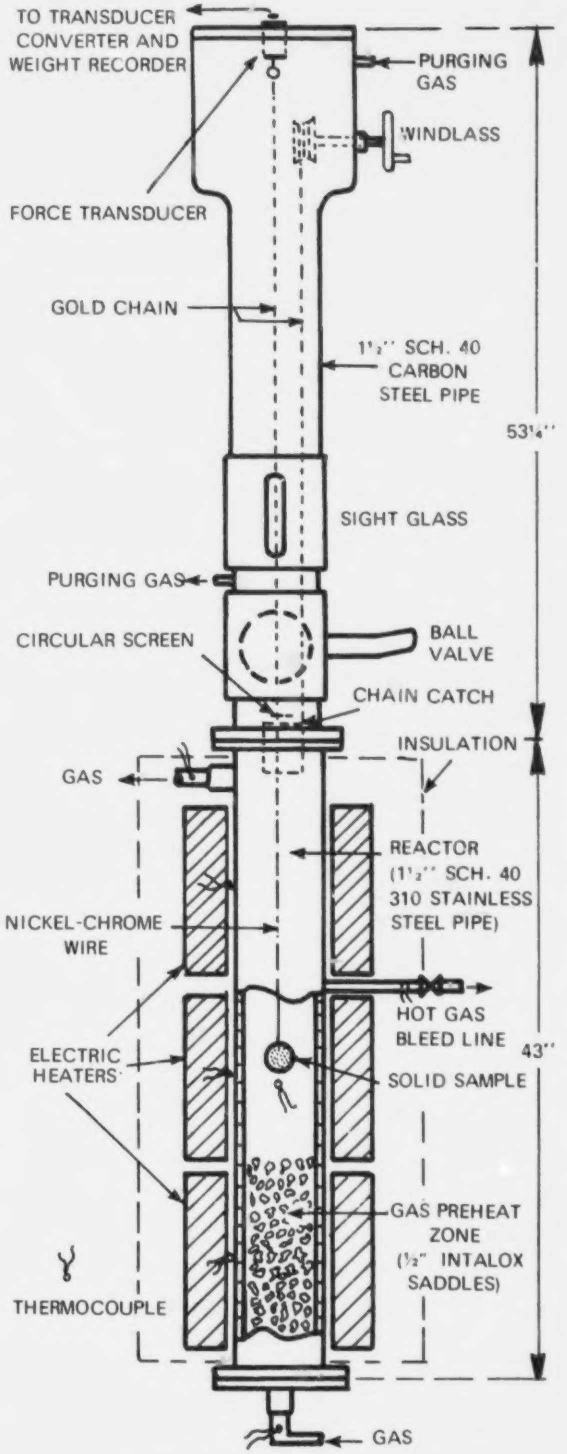

TO TRANSDUCER CONVERTER AND WEIGHT RECORDER
PURGING GAS
WINDLASS
FORCE TRANSDUCER
GOLD CHAIN
1½" SCH. 40 CARBON STEEL PIPE
53¼"
SIGHT GLASS
PURGING GAS
CIRCULAR SCREEN
BALL VALVE
CHAIN CATCH
INSULATION
GAS
REACTOR (1½" SCH. 40 310 STAINLESS STEEL PIPE)
NICKEL-CHROME WIRE
HOT GAS BLEED LINE
43"
ELECTRIC HEATERS
SOLID SAMPLE
GAS PREHEAT ZONE (½" INTALOX SADDLES)
THERMOCOUPLE
GAS

Figure 7. Thermobalance.

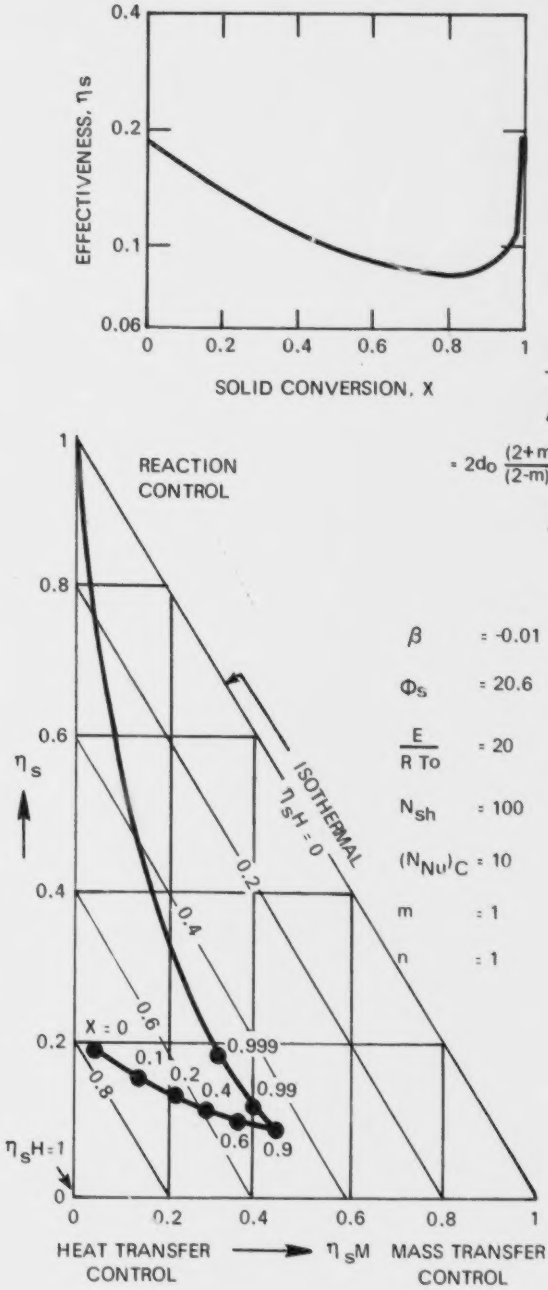


Figure 8. Triangular diagram showing the reaction path and the magnitude of each resistance in an endothermic reaction.4

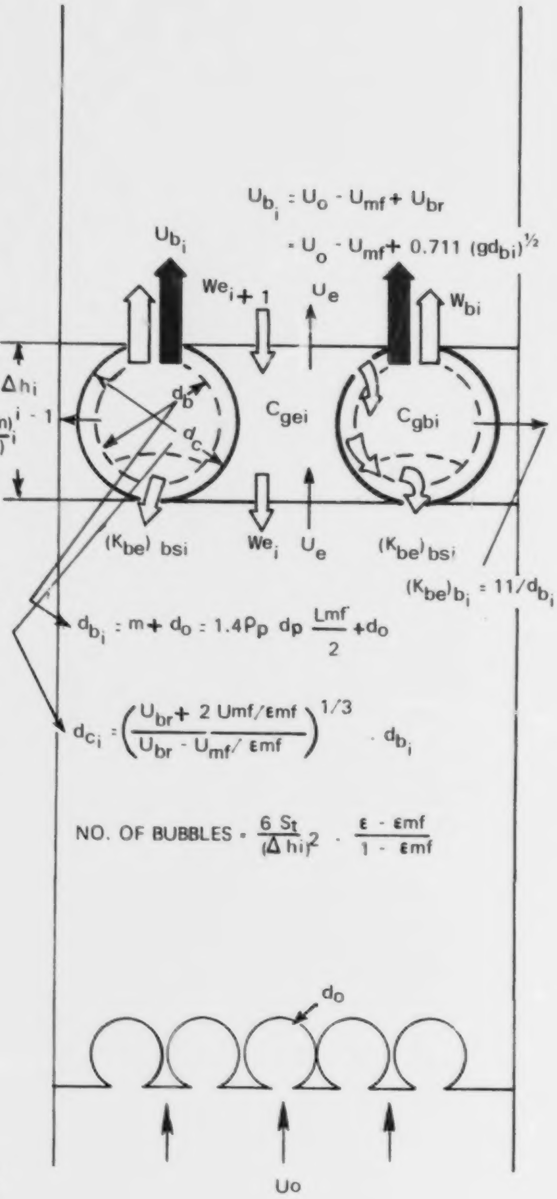

$U_{b_i} = U_o - U_{mf} + U_{br}$
$= U_o - U_{mf} + 0.711\,(g d_{bi})^{1/2}$
U_{b_i}
We_{i+1}
U_e
W_{bi}
Δh_i
$= 2d_o \frac{(2+m)^{i-1}}{(2-m)^i}$
d_b
d_c
C_{gei}
C_{gbi}
$(K_{be})_{bsi}$
We_i
U_e
$(K_{be})_{bsi}$
$(K_{be})_{b_i} = 11/d_{b_i}$
$d_{b_i} = m + d_o = 1.4 \rho_p\, d_p \frac{L_{mf}}{2} + d_o$
$d_{c_i} = \left(\frac{U_{br} + 2\,U_{mf}/\epsilon_{mf}}{U_{br} - U_{mf}/\epsilon_{mf}}\right)^{1/3} \cdot d_{b_i}$
NO. OF BUBBLES = $\frac{6\,S_t}{(\Delta h_i)^2} \cdot \frac{\epsilon - \epsilon_{mf}}{1 - \epsilon_{mf}}$
d_o
U_o

Figure 9. Schematic diagram of i-th compartment in the bubble-assemblage model for fluidized bed.7

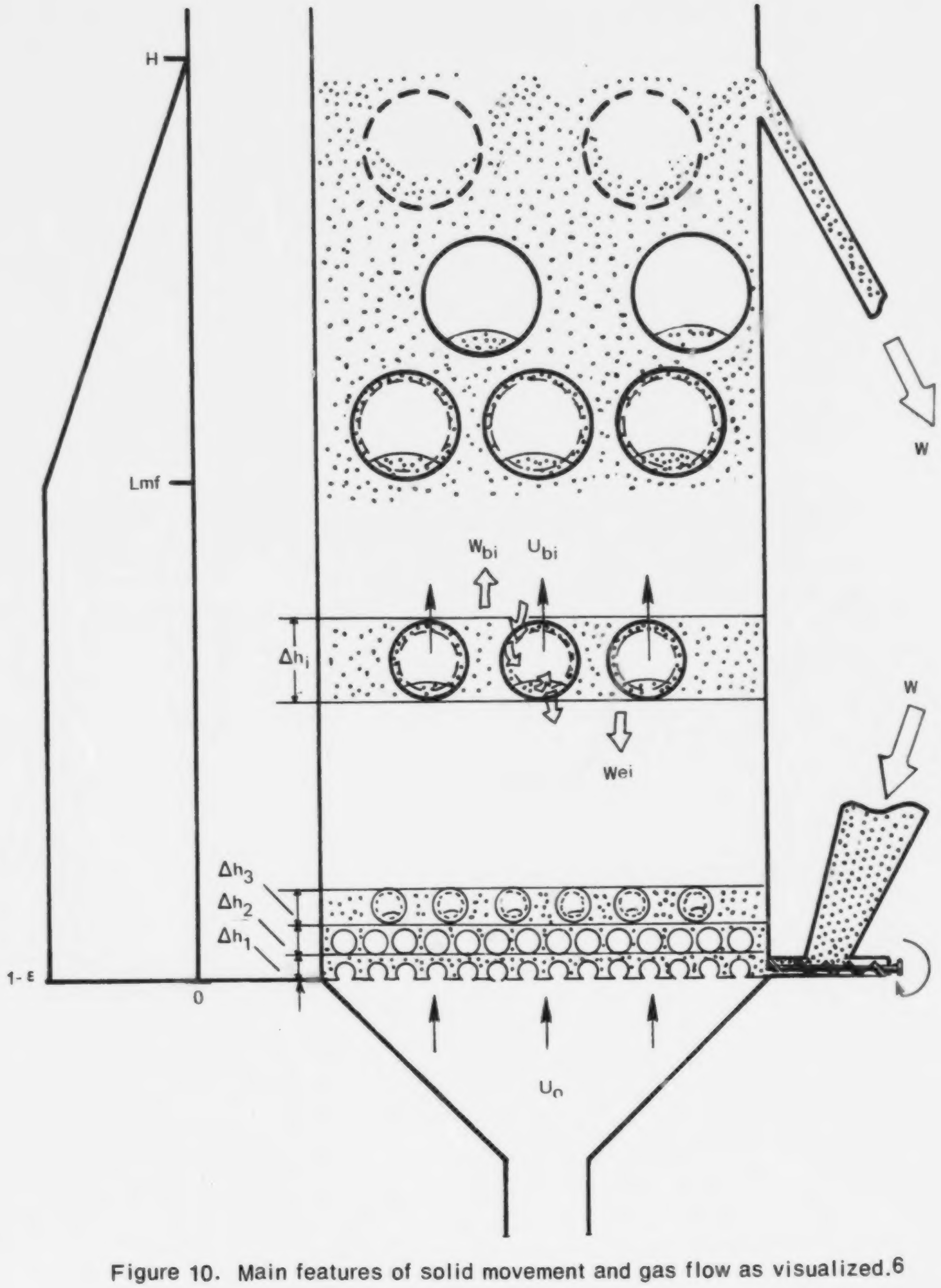

H
Lmf
1- ε
0
W
W_{bi}
U_{bi}
Δh_i
W
Wei
Δh_3
Δh_2
Δh_1
U_o

Figure 10. Main features of solid movement and gas flow as visualized.6

7. MANUFACTURE OF ACTIVATED CARBON BY GASIFICATION IN A FLUIDIZED BED

A. A. GODEL

Societe Anonyme Activit

INTRODUCTION

Today, I propose abandoning momentarily the study of combustion problems in fluidized beds in order to discuss chemically the manufacture of activated carbon.

Activated carbon is a most powerful absorbent. It is used to purify both air (removing malodourous gases) and liquids (purifying water and blanching sugar.)

The principle of the manufacture of activated carbon which concerns us here consists of subjecting carbonaceous matter (e.g., charcoal, peat lignite, coal, and briquettes) to the action of steam at about 800 - 1000°C.

In fact, this treatment represents a true gasification process involving selective oxidation of tarry matter contained in the carbonaceous matter. It is sometimes considered–in a rather simplified manner–as a cleansing operation for existing pores or, for pores created by the operation within the internal structure of the carbon, pores and capillaries which thus become "absorbent."

In 1948, our factory at Vernon was the first in France to manufacture activated carbon by fluidization, in accordance with a continuous-flowing thin layer.[1] This process was later licensed to the French Societe "Carbonisation & Charbons Actifs" which applied it in two industrial furnaces in its factory at Parentis (Landes).

NEW TECHNIQUE

However, it's not about this continuous activation technique which I should like to speak today, but rather about an entirely new technique, still in the experimental design stage, that is not yet developed industrially.

Contrary to the proceding process, the new process is characterized by treating carbonaceous matter in deep fluidized beds under the action of pure stream at high temperature, by discontinuous "batch process" operation.

As indicated previously, this operation results in the gasification of a more or less important fraction of the raw material. To obtain certain grades of activated carbon, it is sufficient to gasify a quarter or a third of this raw material; for others, the process must be carried further–to two-thirds or even three-quarters. The manufacture of activated carbon is thus linked with gasification. Yield, that is the ratio between the weight of the activated carbon obtained and the weight of raw material charged, varies not only according to the nature of the carbon obtained, but also according to the processing methods used: the goal of the new process is to considerably improve technical efficiency.

In general, the production of activated carbon and gasification result from reactions of steam at red heat on carbon; these are pressure- and temperature-dependent balancing reactions situated between two equations:

$$C + H_2O = CO + H_2 \qquad (1)$$

(28.8 K calories per mole)

$$C + 2\,H_2O = CO_2 + 2\,H_2 \qquad (2)$$

(14.8 K calories per mole)

Equation (1) is dominant above 1000°C (1830°F), requiring practically no excess steam; equation (2) is dominant below 800°C (1470°F), requiring, by comparison, steam consumption significantly greater than the stoichiometric ratio. It is for this reason that I have linked here the manufacture of activated carbon with the gasification process. I shall limit my discussion, however, to the manufacture of activated carbon and shall simply mention, in passing, possibilities for the application of this same process to integral fuel gasification.

In this new technique, we operate by batch process and not by thin-layer fluidized-bed continuous flow: we do this to avoid both activated carbon degradation during processing by bituminous gases which are given off, and the inconvenience which occurs inevitably during continuous fluidized flow due to the mixture of crude and activated fractions.

Having adopted the high-temperature batch process, in order to obtain the best weighted yield and best gas production, it is obvious that we must operate in cyclic phases. To obtain, during one reaction phase, the important calorie transfer necessary to maintain the temperature at 1000°C (1830°F), in spite of reaction endothermicity, it was particularly essential to operate by fluidization in order to make maximum use of the excellent heat transfer ratio existing by contact with the internal heating surfaces. In the new process, these heating surfaces will be constituted by fixed heat-accumulating masses preheated to high temperature in a preceding phase.

As you may note, this technique resembles somewhat that of the production of water gas in fixed layer; however, instead of using treated carbon for the accumulation of calories, which would only lead to insignificant phase durations (a few minutes only), we use important fixed heat-accumulating masses which permit extending phase durations to more than half an hour or an hour. Under these circumstances, fluidized-bed temperature may be made to oscillate, at will, around the average reaction design temperature, whether for equation (1) or (2); although each absorbs an unequal, but considerable, quantity of heat, the oscillations are of the order of 1000°C maximum.

It has always been difficult to achieve such heat transfers by traditional processes; e.g., an admixture of flue gas with steam at high temperature, or the use of retorts heated from the outside by flames, thus resulting, in practice, in too low temperatures giving low yield.

The following figures will clarify the question of the required heat supply – the gasification of 12 (the atomic weight) grams of carbon, according to reaction 1 already mentioned as applicable at 1000°C (1830°F), requires the following heat supply:

1. For endothermicity, 28.8 calories.
2. For additional superheating of the steam, between 450 and 1000°C (840 and 1830°F), 6.3 calories.

Although theoretical, these figures show clearly that endothermicity dominates and that it is easy to complete superheating of steam from 450 to 1000°C (from 480 to 1830°F) by direct introduction into the fluidized bed. The steam is produced by the recuperating boiler and introduced into the base of the reactor at precisely 450°C (840°F). To work in cyclic phases, a productional unit will preferablly be provided with two reactors, either coupled or separate, but which operate in alternative phases: that is, one gasification phase and one reheating phase of heat-accumulating refractory masses. It is, of course, possible to have production units consisting of three or more reactors.

Each reactor is equipped with piles of refractory material forming heat-accumulating masses which will be placed in good contact with the fluidized bed. These masses will be chosen on the basis of their refractoriness, their non-adherence of clinkers, and their good thermal conductivity.

With regard to thermal conductivity, certain carborundum agglomerates which are highly refractory (1650 - 1700°C–3000-3100°F) have thermal conductivity 5 or 10 times greater than that of current refractory materials, or almost equal to that of stainless steel.

TWO EXAMPLES

To illustrate the method of construction and operation of the new process, I shall describe two examples, although other combinations may be envisaged to correspond to various uses.

Twin-Reactor Furnace

In the first example, represented by Figures 1 and 2, the two reactors in the activated carbon and gasification production unit are juxtaposed in a single rectangular furnace divided into two identical, but distinct, parts (A and B); each reactor (A and B) contains a nest of parallel, vertical, heat-accumulating refractory slabs (1). These slabs swell out at the base (2), leaving only a narrow slot (3) used for introducing reaction steam; the fluidized bed is supported by this steam injection during the entire blowing period which begins when carbonaceous material is charged into reactor A at the beginning of the reheating phase. The material is charged pneumatically at (4), reaching a level slightly higher than that of the slabs.

At the end of the reheating phase, the steam is stopped, thus provoking instantly the flow of carbon by (3) in the hopper. The activated-carbon product is evacuated pneumatically by (5) toward storage (not shown).

During the reaction (or gasification) phase in reactor A, where gas is produced, air is introduced at (6′) in reactor B; this gas thus burns in the upper chamber of reactor B which is then in reheating phase. The combustion gases reheat the slabs (1′) and escape at the base at (3′). They are finally evacuated from the reactor by (7′) to a recuperating waste heat boiler (not shown) which produces superheated steam at 450°C (840°F) for activation; this superheated steam is introduced in reactor A at (8). Slab thickness, usually greater than their spacing, is calculated so as to obtain required calorific capacity corresponding to phase and reacting duration.

The set of reactors A and B are put into operation quite simply: when empty, they are reheated simultaneously to service temperature by gas or fuel oil burners (9) and (9′). When a temperature of about 1200°C (2190°F) is reached in the reactors and when the boiler is under pressure, the burners are stopped and the set of reactors is placed in cyclic reversal operation by injecting, at the beginning of the phase, alternately in one reactor then in the other, the carbonaceous material to be activated. The steam must be injected from the beginning, in the lower part, and during the active charging and gasification operations.

Cylindrical Double-Reactor System

In the second example, illustrated by Figures 3 and 4, the two reactors A and B are separate and cylindrical, permitting the use of cyclone dust collectors for treating gases given off during reaction, and also operation under a pressure of several bars.

The heat-accumulating mass placed in each reactor (A and B) is composed of a refractory block (1), run through by a nest of 20 cm diameter tubes (2). These tubes narrow out at the base so that the orifice (3) at their ends may let pass the reaction steam injected through (8); the steam backs up the fluidized bed which has been previously charged into reactor A to a level slightly higher than that of the heat-accumulating block.

Steam blow-in through (8) must begin in reactor A at the very beginning of the phase, just before introducing the carbonaceous material pneumatically through (4); it is stopped at the end of the activation-reaction phase when the carbon flows down immediately, to be evacuated pneumatically through (10) toward storage (not shown).

Reaction-gas dust collecting takes place in a cyclone (12) in which the lightest particles are deposited; these particles, consisting mainly of activated carbon, may be either reinjected into reactor A or collected directly (as shown).

The gas thus treated is fed through (11) into a combustion chamber (15) where air is introduced through (14); the emitted flue gas is introduced through (16′) to the bottom of reactor B and, therefore, during the next phase through (16) to reactor A; flue gas thus passes from the bottom to the top of reactor B and through a bed of finely granulated refractory material–on top of the nest of tubes in the accumulating block–introduced pneumatically through (6′) at the beginning of the reheating phase. Obviously, this granulated material contributes no calories of itself, but constitutes an auxiliary fluidized bed that increases the transmission of calories to the walls of the nest tubes during reheating. The granular refractory material may be a sulfur acceptor.

The flue gas evacuated from reactor B passes through cyclone (13) which retains the flue dust eventually emitted from the refractory material. Flue gas is finally evacuated to a waste heat boiler, not shown.

At the end of the reactor B reheating period, which coincides with the end of the reactor A activation period, gasification is stopped and so is the circulation of flue gas. The refractory material then flows freely down through the lower orifices of the tubes, from where it is evacuated at the base through (10′) and conveyed pneumatically into a silo (not shown) where it awaits transfer during the next reactor A heating phase. When reactor B is reheated, it is ready to receive first the steam, and then the load of carbonaceous material: the cycle thus continues.

System Operation

To put reactors A and B into operation, procedure is as indicated in the first example (Figures 1 and 2): they are heated simultaneously by gas or fuel oil burners at (9) and (9′).

In both described installations, air and steam valves are ordinary, but the valves for evacuating activated carbon, gas, and flue gas are water cooled.

Valves operate automatically in accordance with programming which takes into account heating requirements. These requirements may vary considerably according to the nature of the carbons treated, or rather, according to the nature of the ash of these carbons: if the ash is extremely fusible, it may be evacuated in melted form, in which case, it would be advisable to adopt a high reaction-temperature solution. If the ash is not extremely fusible, temperature levels lower than ash melting point will be used and the ash will be evacuated in powder form together with the activated carbon: ash will then be separated, using commonly used industrial processes: e.g., dust screening and washing.

Coming back to the second type of installation which I have just described (Figures 3 and 4), the following figures are provisional estimates.

The inside dimensions of the unit, illustrated by Figure 4 and which corresponds to a production capacity of 6 to 12 tons per day (even 18 tons per day, for production varies considerably with the quality of the activated carbon needed) are:

1. Section of each cylindrical reactor, 6 sq m.
2. Inside diameter, 2.80 m.
3. Height of the cylinder, 3.50 m.
4. Total height of heat-accumulating refractory material in each reactor, 1 m.
5. Total volume of heat-accumulating refactory material in each reactor, 4 cu m.

Production of superheated steam by the waste heat boiler is superabundant; for this reason, excess gas can be tapped through (17).

Operation under a pressure of several bars may be desirable, either in increase the production of activated carbon or to improve the purification of the gas in view of its use in gas turbines.

Another way of increasing the production of activated carbon and gas is to operate with a highly expanded fluidized bed.

In fact, it is conceivable that if the installation of the reactors must be adapted to integral fuel gasification, the compressed gas produced, perfectly desulphurized, might be used in turbines for the production of energy in a mixed- gas-steam cycle, without atmospheric pollution.

CONCLUSION

I must state, however, that the process which I have just described to you was specifically designed for the production of activated carbon, with the simple aim of being able to adapt it eventually to integral fuel gasification for the production of synthesis gas, rich in hydrogen and carbon monoxide, without either nitrogen or carbon dioxide. The gas produced, being of a highly reducing nature, should be easily desulphurized by traditional scrubbing methods or by absorption in dry conditions by sulfur acceptor, according to new processes such as Professeur Squire's, thus permitting effective recuperation of pure sulfur.

In conclusion, I should like to state that activated carbon, the subject of the manufacturing process which I have just described, is one of the most precious auxiliaries for the protection against both atmospheric and water pollution agents.

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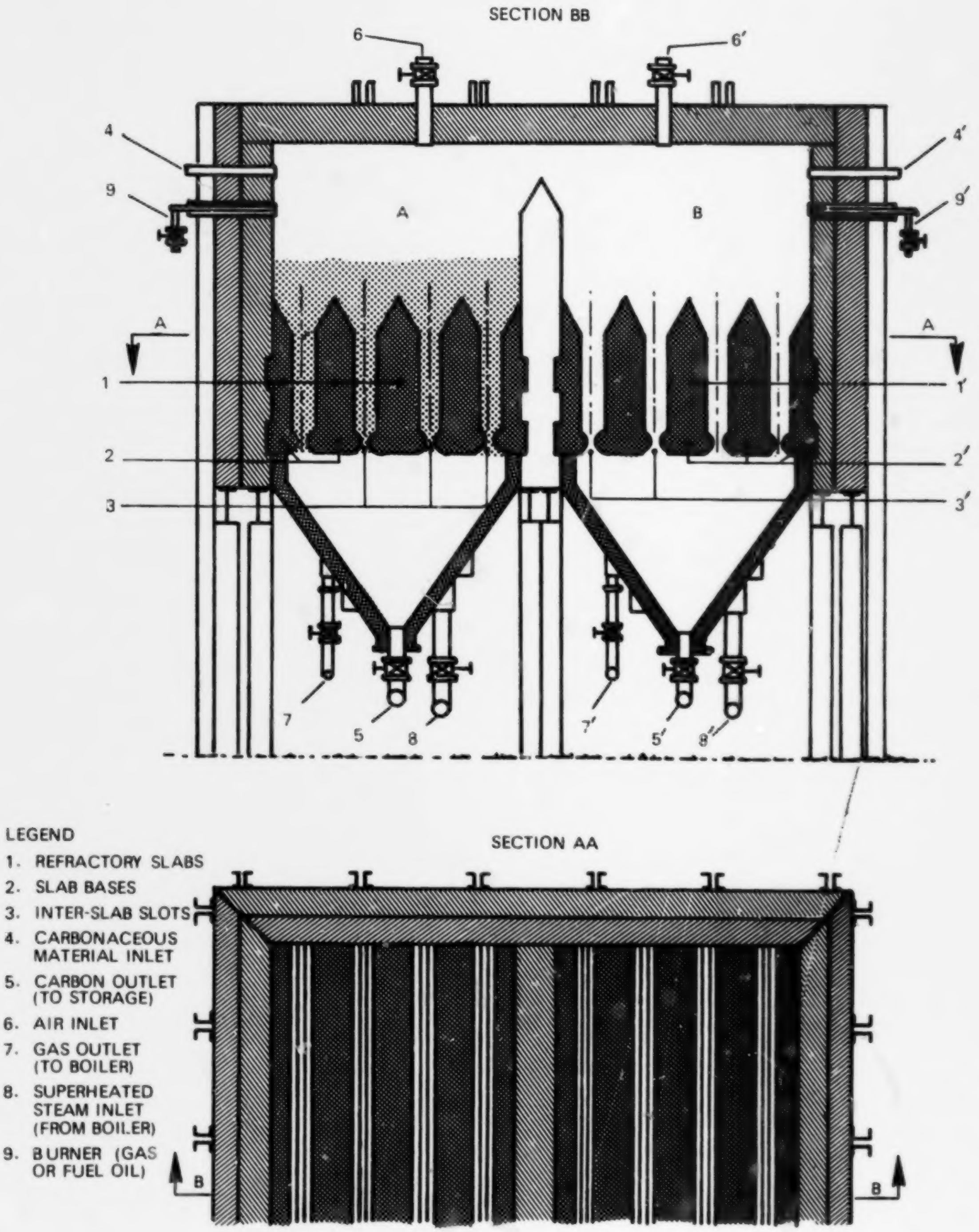
2. Swiss patent No. 250, 891.

3. French patents No. 942, 699 and 951, 153.


SECTION BB
6
6′
4
4′
9
9′
A
B
A
A
1
1′
2
2′
3
3′
7
5
8
7′
5′
8′
SECTION AA
B
B

LEGEND
1. REFRACTORY SLABS
2. SLAB BASES
3. INTER-SLAB SLOTS
4. CARBONACEOUS MATERIAL INLET
5. CARBON OUTLET (TO STORAGE)
6. AIR INLET
7. GAS OUTLET (TO BOILER)
8. SUPERHEATED STEAM INLET (FROM BOILER)
9. BURNER (GAS OR FUEL OIL)

Figure 1. Twin-reactor furnace.

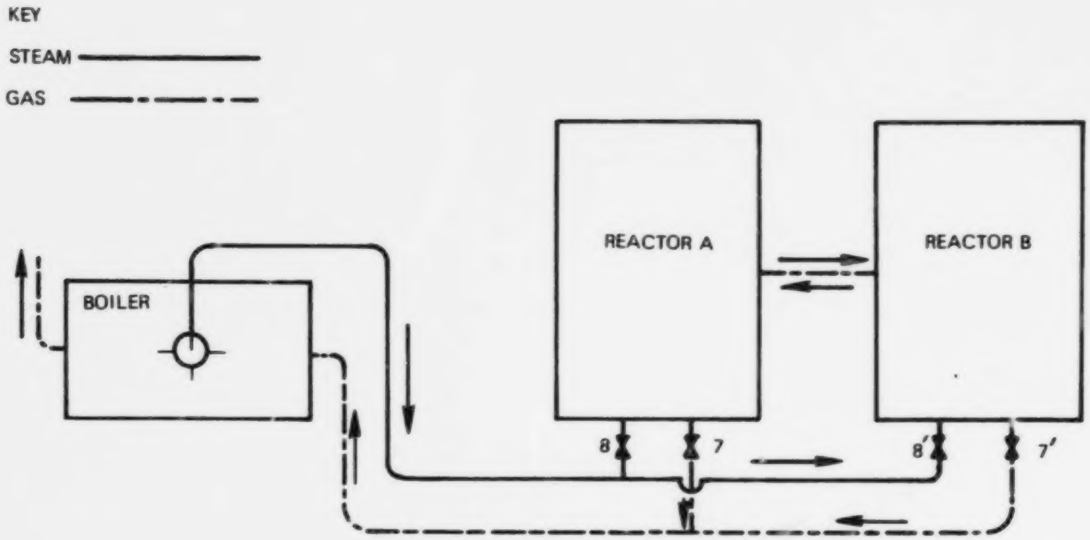

KEY
STEAM
GAS
REACTOR A
REACTOR B
BOILER
8
7
8'
7'

Figure 2. Twin-reactor furnace, flow sheet.

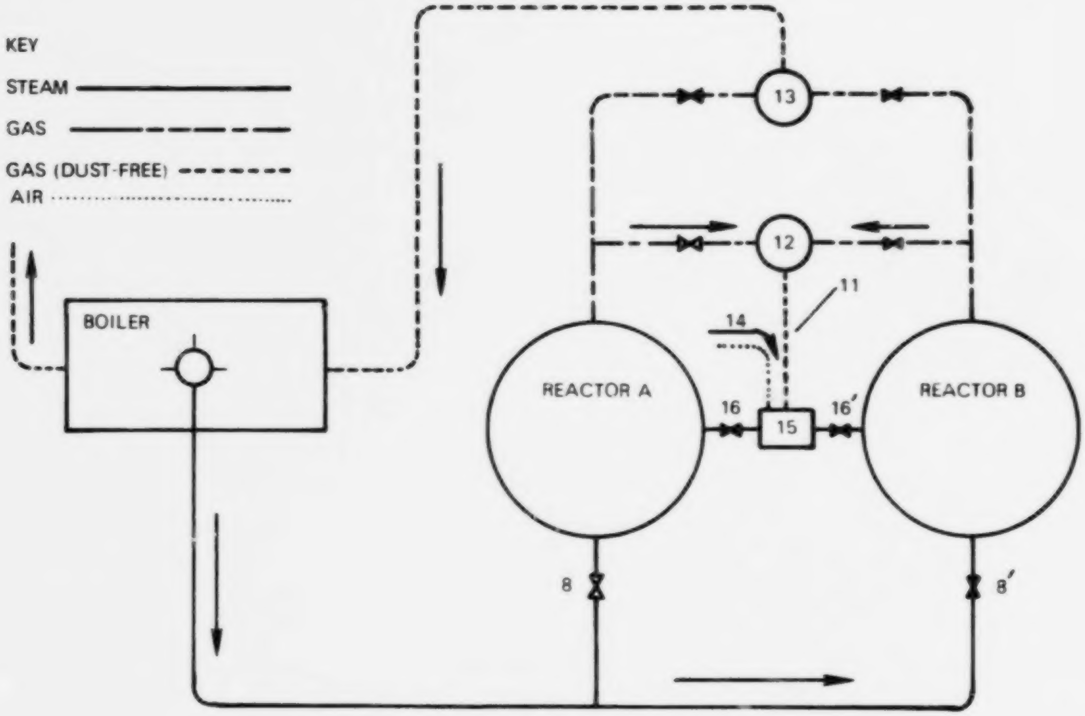

KEY
STEAM
GAS
GAS (DUST-FREE)
AIR
BOILER
13
12
11
14
15
16
16'
REACTOR A
REACTOR B
8
8'

Figure 3. Cylindrical double-reactor system, flow sheet.

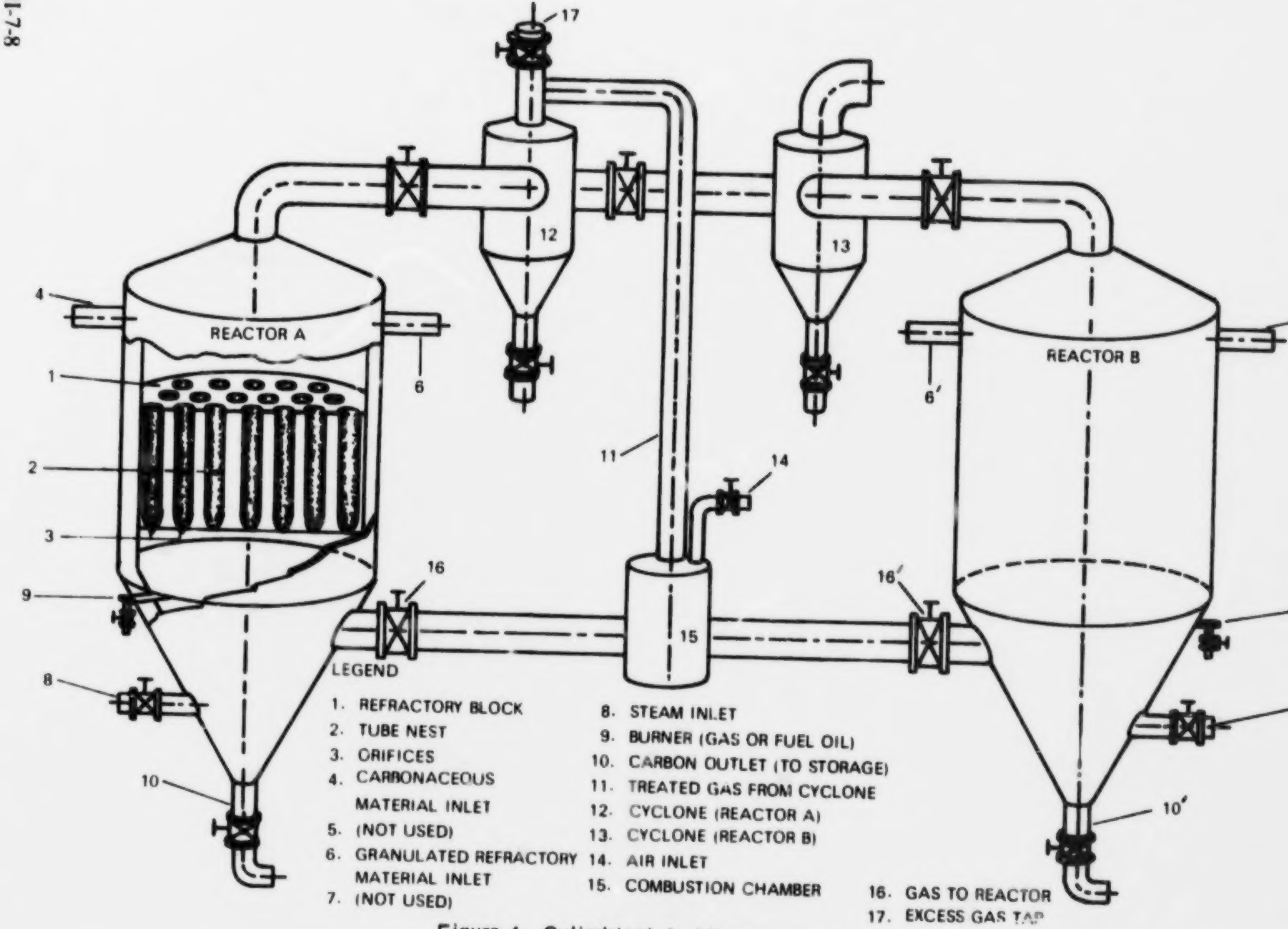

17
12
13
4
REACTOR A
6
1
2
3
9
8
10
11
14
16
15
16'
4'
REACTOR B
6'
9'
8'
10'
LEGEND
1. REFRACTORY BLOCK
2. TUBE NEST
3. ORIFICES
4. CARBONACEOUS MATERIAL INLET
5. (NOT USED)
6. GRANULATED REFRACTORY MATERIAL INLET
7. (NOT USED)
8. STEAM INLET
9. BURNER (GAS OR FUEL OIL)
10. CARBON OUTLET (TO STORAGE)
11. TREATED GAS FROM CYCLONE
12. CYCLONE (REACTOR A)
13. CYCLONE (REACTOR B)
14. AIR INLET
15. COMBUSTION CHAMBER
16. GAS TO REACTOR
17. EXCESS GAS TAP

Figure 4. Cylindrical double-reactor system.

SESSION IV:

Conceptual Design and Economic Feasibility

SESSION CHAIRMAN:

Dr. D.H. Archer, Westinghouse

1. CLEAN POWER SYSTEMS USING FLUIDIZED-BED COMBUSTION

A. M. SQUIRES

The City College of The City University of New York

INTRODUCTION

A major opportunity for clean power from coal involves the combustion of coal at high pressure in presence of a desulfurizing agent, and the generation of power by a combination of gas- and steam-turbine cycles.

This paper reviews briefly three candidate combustion technologies for use in the foregoing combination, and considers power cycle arrangements suited for each of the three approaches to combustion.

HIGH-PRESSURE SULFUR CAPTURE

In a system for combustion at high pressure, an agent derived from dolomite may advantageously capture sulfur during a first coal-processing step. For this first step there are three cases to consider.

1. Complete combustion, using air in excess of stoichiometric.
2. Partial combustion, using between about a third and half of the stoichiometric air and yielding a fuel gas containing CO and H_2.
3. A carbonization yielding low-sulfur coke as well as fuel gas, with heat for the carbonization supplied by a partial combustion which consumes on the order of 10 percent of the stoichiometric air.

Work is underway at The City College on the kinetics of three reactions whereby gaseous sulfur species may be absorbed by agents derived from dolomite:

$$[CaCO_3+MgO] + SO_2 + 0.5\ O_2 = [CaSO_4+MgO] + CO_2 \qquad (1)$$

$$[CaCO_3+MgO] + H_2S = [CaS+MgO] + H_2O + CO_2 \qquad (2)$$

$$[CaO+MgO] + H_2S = [CaS+MgO] + H_2O \qquad (3)$$

Our primary interest is in the application of the reactions in systems where the solid is used cyclically, a step for desorption of H_2S from the solid being present in the cycle, thus:

$$[CaS+MgO] + H_2O + CO_2 = [CaCO_3+MgO] + H_2S \qquad (4)$$

The equilibrium for reaction (4), the reverse of reaction (2), is a strong function of temperature; formation of H_2S is favored by a lower temperature. To obtain a gas containing H_2S at a concentration suitable for economic conversion to sulfur in a Claus system, reaction (4) should be conducted at a temperature in the general vicinity of 600°C and at elevated pressure.

This paper is confined to a consideration of each of the three aforementioned combustion technologies in a version which employs an appropriate reaction selected from (1), (2), and (3) for the capture of sulfur, along with reaction (4) for the release of sulfur from the solid.

Complete Combustion Approach

Hoy and Stanton[1] have described a large-scale experiment for fluidized-bed combustion of coal at 6 atmospheres and 800°C. They propose a pressure of 15 atmospheres for the commercial embodiment of this technique. About 70 percent of the heating value of coal would be transferred to boiler tubing within the bed; the remaining 30 percent would be available as sensible heat in the 800°C effluent gas, which would be expanded in a gas turbine. The authors hope that alkali and dust content of the gas will be acceptable for its direct use in the turbine, so that equipment need not be provided to cool the gas, scrub it of alkali dust, and reheat the gas to a suitable turbine-inlet temperature.

The CO_2 partial presure in gas leaving the complete-combustion fluidized bed is sufficiently great that $CaCO_3$ in dolomite added to the bed would not decompose. Half-calcined dolomite present in the bed would react readily with SO_2, as in reaction (1). Kinetics for this reaction, apparently on account of half-calcined dolomite's porosity, are favorable at temperatures even as low as 600°C.[2,3,4]

The team at The City College has demonstrated that a way exists to regenerate $[CaCO_3+MgO]$ from $[CaSO_4+MgO]$ while liberating H_2S for sulfur manufacture.[4] The sulfate would be reduced to sulfide at about 800°C, thus:

$$[CaSO_4+MgO] + 4\,H_2 = [CaS+MgO] + 4\,H_2O \qquad (5)$$

The sulfide would be converted to the carbonate by reaction with steam and carbon dioxide at about 600°C, as in reaction (4). Reactions (5) and (4), as well as reaction (1), would be conducted at elevated pressure, suitably the 15 atmospheres which Hoy and Stanton proposed.

In our recent paper,[4] we offered a scheme employing methane reforming as a source of hydrogen for reaction (5). In the context of Hoy and Stanton's proposal, reaction (5) might advantageously be carried out in a small auxiliary fluidized bed operating under reducing conditions. Solid would be transferred from the main fluidized-bed boiler to the auxiliary bed and thence to a fluidized bed for sulfur desorption by means of reaction (4).

It is important to note that the maximum temperature for the dolomite-derived solids would be 800°C in this scheme. There is evidence[5] that $[CaO+MgO]$ does not sinter and lose surface area at this temperature. We are examining the question of the sintering of other dolomite-derived solids, and have grounds to hope that none of the solids will undergo serious sintering under this complete combustion approach. We would expect solid reactivity to remain excellent through a large number of cycles. The practical importance of this fact will, of course, depend upon one's degree of success in separating the dolomite-derived solid from coal ash to be discarded.

Partial Combustion Approach

If partial combustion of coal were conducted in a fluidized bed of fully-calcined dolomite, sulfur would be absorbed by reaction (3). Reaction (4) could then be used to desorb sulfur. The resulting carbonated solid, $[CaCO_3+MgO]$, could be returned to the partial combustion bed, to be calcined therein.

If all of the carbon in coal is reacted with oxygen in air to form CO, a surplus of heat is developed. The surplus heat could be used to raise steam in boiler tubes passing through the partial combustion bed or to heat combustion air. Alternatively, heat from the formation of CO could be used to support the endothermic reaction of steam with carbon, steam being supplied to the bed along with air. Adding steam to the bed would, of course, increase the latent heat loss from the overall power-generating system. This is not quite as bad as might appear at first glance. Steam supplied to the partial combustion step will eventually reappear as steam at the gas turbine, and the total flow of gas through the turbine will be larger than if steam had not been supplied to the partial combustion step. This means a larger net production of power from the gas turbine, and the cost of the gas turbine, including air compressor, will be significantly less than if steam were not used. This reduction in capital cost will provide an economic offset against the reduction in efficiency chargeable to the latent heat loss. The higher the gas-turbine inlet temperature and the higher the pressure, the less advantageous it will be to use the excess heat from partial combustion to raise steam for a steam cycle and the more advantageous to use this heat to decompose steam by reaction with carbon.

Partial combustion has potential advantages over complete combustion:

1. About half as much gas would be produced, and so the fluidized bed would be smaller.
2. The gas, containing potential energy as well as sensible heat, would represent a larger proportion of the heating value of the coal and would permit a second combustion step in which the gas would be heated to a much higher temperature at the inlet of the gas turbine.

The drive for higher performance in aircraft engines will continue, and experience from such engines can be expected to maintain the historic upward trend of temperature of gases at the inlet of industrial gas turbines. As this temperature rises, the second advantage of partial combustion may appear progressively more important.[6]

Partial combustion has difficulties, however:

1. Sufficient carbon must be present in the bed to support the CO_2-carbon and H_2O-carbon reactions. Carryover from the bed will contain carbon which must probably be burned elsewhere, perhaps best in an auxiliary bed for complete combustion.
2. The temperature must be significantly higher, probably at least 900°C, to cater to the kinetics of the CO_2-carbon reaction. The higher temperature increases the danger that alkali in the gas will injure the gas turbine, and therefore increases the likelihood that gas will have to be cooled and scrubbed.

If gas must be cooled and scrubbed to deal with alkali from partial combustion, but not from a total combustion at 800°C as Hoy and Stanton hope, then the advantages of partial combustion might disappear in a total economic balance, at least for gas-turbine inlet temperatures currently available.

When a temperature is selected for the partial combustion bed, attention must be paid to the problem of sintering of the dolomite-derived solid. Preliminary work at The City College confirms Haul's finding[5] that some sintering of [CaO+MgO] occurs at temperatures in the vicinity of 925-975°C. There are indications, however, that the solid stabilizes at a surface area on the order of about 5 m^2/gram. It must be emphasized that this is a preliminary conclusion, but it would appear reasonable to expect that a temperature below about 975°C should be compatible with a relatively long life for the dolomite-derived solid in a partial combustion bed.

If the gases from such a bed must be cooled and scrubbed, there may be an advantage of an ash-agglomerating fluidized bed operating at a higher temperature and not incorporating an agent to capture sulfur. Gases from the bed at higher temperature will be lower in H_2O and CO_2 content, and hence will be admirably suited for desulfurization by reaction with half-calcined dolomite, as in reaction (2). The gases should be cooled several hundred degrees before the desulfurization step. We have been surprised to discover that reaction (2) is considerably faster, at a given temperature and H_2S partial pressure, than reaction (3). Accordingly, a panel bed filter[7] is well suited for carrying out reaction (2). Perhaps such a filter, with the help of a filter aid, can remove alkali dust from the gas to meet the cleanliness required for the gas-turbine inlet. It will be important to learn whether or not this is so. The filter could operate at a temperature below 800°C, and the life of the filter solid should be excellent.

Carbonization Approach

One of the great inventions is the proposal by Gorin, Curran, and Batchelor[8] to desulfurize coal char by cooperative action of hydrogen and an "acceptor" for sulfur, such as calcined dolomite. Under the proposal, the acceptor and coal char would differ sufficiently in particle size and density so as to be readily separable.

This proposal's originality is appreciated if it is recalled that the scientific basis for the idea was available as early as 1923 from the work of Powell[9] of the U.S. Bureau of Mines.

The inventors' apparent major interest was to provide a low-sulfur char for use in making "formcoke" for metallurgy.

Theodore[10] elaborated the idea to produce a low-sulfur coke for power generation while converting volatile matter in the coal to pipeline gas.

I have examined the idea's potential for a system intended simply to provide low-sulfur fuels for power.[11,12,13] Figure 1 depicts broadly a scheme we are studying. The coal would be carbonized at elevated pressure and the resulting coke desulfurized by cooperative action of hydrogen and the acceptor. Heat for the distillation of volatile matter from coal would be supplied by partial combustion of the volatile matter, typically consuming 11 percent of the stoichiometric air for complete combustion of the coal. Hence, equipment volumes would he even less than in the partial combustion approach. The quantity of lean fuel gas is relatively small, and equipment for its cooling and scrubbing, if necessary, would be correspondingly small. The lean fuel gas would drive a gas turbine, and a satellite steam power station would receive waste heat from the gas turbine's exhaust as well as from the coal desulfurization process. An installation would operate at a steady coal feed and would provide baseload power corresponding to about a third of the heating value of the coal. The installation would furnish low-sulfur coke for production of variable power, either in nearby equipment or at a distance.

POWER PRODUCTION FROM LOW-SULFUR COKE

I will defer a discussion of the scheme itself, and first take up alternatives for the production of power from the low-sulfur coke.

Alternative 1

The coke might be burned in existing power stations. Grinding the coke for pulverized boilers might be costly, but using the coke in stations having cyclone combustors might be relatively easy, if the coke were made from coal having ash suitable for such combustors. Some gaseous or volatile fuel might be needed to support combustion of the coke, but many utilities have now switched to low-sulfur oil, and would welcome the opportunity to replace even a part of this expensive oil with a cheaper fuel. A modification of Figure 1 might provide a suitable gaseous or liquid fuel produced from the coal itself.

Alternative 2

Elliott[14] has pointed out the advantages of the fluidized-bed boiler for stations whose purpose is to meet peakload requirements. The coke would be an excellent fuel for such stations, relieving their owners of the necessity of providing equipment to cope with sulfur. It would appear that the cost advantage might belong to a design at high pressure, such as that proposed by Hoy and Stanton.

Alternative 3

The efficiency of Hoy and Stanton's scheme would be improved if gas from the fluidized-bed boiler were heated to a higher temperature ahead of the gas turbine by a combustion of fuel derived from volatile matter. This idea is depicted broadly in Figure 2. The figure assumes, perhaps too pessimistically, that the gas from the fluidized-bed boiler must be cooled and scrubbed to remove dust. If this pessimistic assumption is correct, the availability of a gaseous fuel to reheat the boiler offgas would be welcome.

Alternative 4

The coke might be gasified in a partial combustion process, providing fuel gas to drive a gas turbine. A wider range of alternative gasification processes might be considered for the low-sulfur coke than for raw coal. For example, a process using an ash-agglomerating fluidized bed might seem more attractive for the low-sulfur coke than for coal. If the coke has a sufficiently large particle size, it would be a good fuel for Secord's slagging, gravitating-bed gasifier.[15,16]

This alternative becomes attractive if a major part of the coke is to be burned to provide baseload power, and if advantage is to be taken of the higher gas-turbine inlet temperatures expected to become available.

Alternative 5

The coke might be burned in a fluidized-bed boiler using total combustion and heating steam to temperatures well beyond those now in use in conventional power equipment. Elliott[17] has called attention to this opportunity. In light of present knowledge, development of a device for partial combustion of coal or coke must be viewed as more uncertain than development of a fluidized bed for total combustion. It must also be admitted that the expectation that "ultra-high" gas-turbine inlet temperatures (i.e., beyond 1100°C) will become available is probably not quite yet a certainty. Under these circumstances, it would be a mistake to overlook the opportunity which the fluidized-bed boiler offers to improve steam cycle efficiency by raising the steam temperature.

If the historical line of development of the steam cycle is followed, even a modest increase in steam temperature will be accompanied by a sharp increase in steam pressure. I have called attention to a steam cycle employing an unusually large amount of super heat and affording an efficiency comparable to that of the conventional steam cycle for a given maximum steam temperature, but not requiring such high pressures.[11,18,19] Detailed cost examination is still needed to judge the worth of the new cycle, which should not be altogether overlooked when considering higher steam temperatures.

ACCEPTOR TECHNIQUE FOR DESULFURIZING COAL

Powell's historic work[9], confirmed and extended by Batchelor, Gorin, and Zielke,[20] teaches that the acceptor technique for desulfurizing coal char will work best if the char is carbonized quickly and moved promptly from the carbonization step to the desulfurizing environment. This fact is easily understood when it is remembered that cokes sinter exothermically at temperatures above about 600°C. If the coal char is allowed to sinter, sulfur is locked into the coke structure, the sulfur is inaccessible to reaction with hydrogen, and "deep desulfurization" of the coke is impossible. In thermodynamic terms, sulfur in a coke soaked for a long time at a high temperature is characterized by a free energy such that the pseudo-equilibrium between the sulfur and hydrogen is highly unfavorable for production of H_2S.

The agglomerating tendencies of bituminous coal have posed problems for designers of processes to carbonize such coals in fluidized beds. Many American coals, particularly in the East, are so strongly caking that attempts to carbonize the coals in a simple one-step fluidized-bed carbonization process have often ended in failure caused by the formation of massive agglomerates. Processes have been devised to avoid massive agglomerates by providing several fluidized carbonization stages at progressively higher temperatures. Other processes would inject raw coal, at a relatively low rate, either into a dilute stream of hot recycled char or into a mechanically mixed mass of such char. Either dodge is poor: the first because of the length of time during which the char is soaked at high temperature; the second because of the danger of back reactions returning sulfur to the recycled char.

In an acceptor desulfurization process, there will be an advantage if the coal is ground quite fine, so that desulfurization is not hindered by diffusion of the gaseous reactants, hydrogen and hydrogen sulfide.

Figure 3 is a fuel desulfurization scheme we are studying. Coal is supplied in a finely divided condition, but coke particles are allowed to grow large in a coke-self-agglomerating fluidized bed. Because the agglomerated coke is almost completely nonporous, each fresh patch of coke (produced by capture of a tiny sticky particle of coal) must be desulfurized promptly after it forms upon a coke pellet. To accomplish this, there is a rapid exchange of coke between the agglomerating zone and a desulfurizing zone superposed upon the agglomerating zone. The desulfurizing zone is a fluidized bed of finely divided calcined dolomite. The fluidization velocity in the agglomerating zone is too high to permit the dolomite acceptor particles to sink deep into this zone, and the fluidization velocity in the desulfurizing zone is too low for coke pellets ejected into this zone to remain there permanently.

Because of the small size of the acceptor particles, they may advantageously be calcined in the "recirculating fluid bed" described recently by Reh.[21] Such a bed is indicated schematically for the calcination zone of Figure 3.

Partial combustion of volatile matter supplies heat to calcine the spent acceptor. Partial combustion not only provides a lean fuel gas to drive a gas turbine but is also advantageous because the temperature in the acceptor calcination zone may be lower, for a given total operating pressure, than the temperature which must be specified if complete combustion of a fuel is used to provide heat to this zone. The maximum temperature necessarily reached by the solid in the calcination zone of Figure 3, for operation at 21 atmospheres, would be in the vicinity of 950°C. Acceptor reactivity and life should be good.

Back reactions can restore sulfur only superficially to the desulfurized coke product, for coke pellets produced in a coke-self agglomerating bed will be almost completely nonporous and hence only superficially in contact with gases present in the carbonization step.

Only a pilot plant can establish with certainty the operability of the coke-self-agglomerating zone of Figure 3. We are conducting fluidization tests at atmospheric conditions which are beginning to elucidate the principles governing behavior of a fluidized bed of large particles coated with a sticky substance. The work indicates that such a bed can carry an astonishingly large burden of sticky matter. This result, viewed together with a number of successes elsewhere in the operation of agglomerating beds, makes us confident of the operability of the coke-self-agglomerating zone.

CONCLUSION

The reader may now object that this paper has presented too many proposals, and he may wonder what I consider the best to develop. I would reply that paper research is cheap and the importance of the subject justifies detailed engineering study and cost evaluation of almost any plausible idea which offers itself. The needs of the power industry are so varied that a number of systems may prove commercially viable.

Without such engineering studies, a choice among the systems must be largely subjective, and I can make it only by asking myself, on which systems would I be happy to devote substantial amounts of time and energy? I would, today, pick three systems.

1. The acceptor desulfurization scheme of Figure 3; it offers the overwhelming advantage of providing low-sulfur fuel to existing plant in a form which can be stored and shipped.
2. A bed for complete combustion, either of coal in presence of half-calcined dolomite or of low-sulfur coke from the acceptor desulfurization process.
3. An ash-agglomerating partial combustion bed followed by a panel bed filter at lower temperatures, with the bed consuming either coal or low-sulfur coke; if coal is used, the panel would be charged with half-calcined dolomite.

ACKNOWLEDGEMENTS

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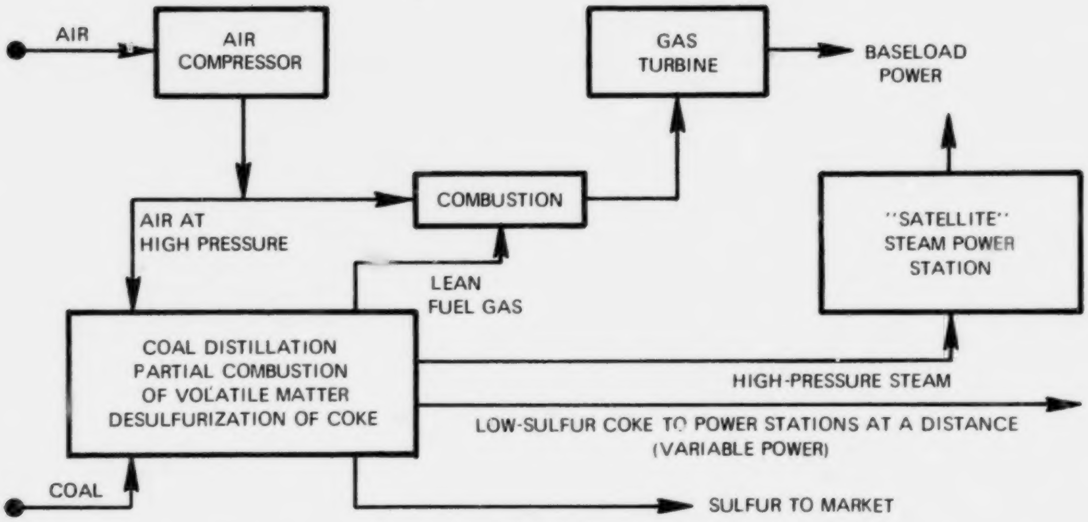
AIR
AIR COMPRESSOR
GAS TURBINE
BASELOAD POWER
COMBUSTION
AIR AT HIGH PRESSURE
"SATELLITE" STEAM POWER STATION
LEAN FUEL GAS
COAL DISTILLATION PARTIAL COMBUSTION OF VOLATILE MATTER DESULFURIZATION OF COKE
HIGH-PRESSURE STEAM
LOW-SULFUR COKE TO POWER STATIONS AT A DISTANCE (VARIABLE POWER)
COAL
SULFUR TO MARKET

Figure 1. Scheme for providing low-sulfur fuels for power.

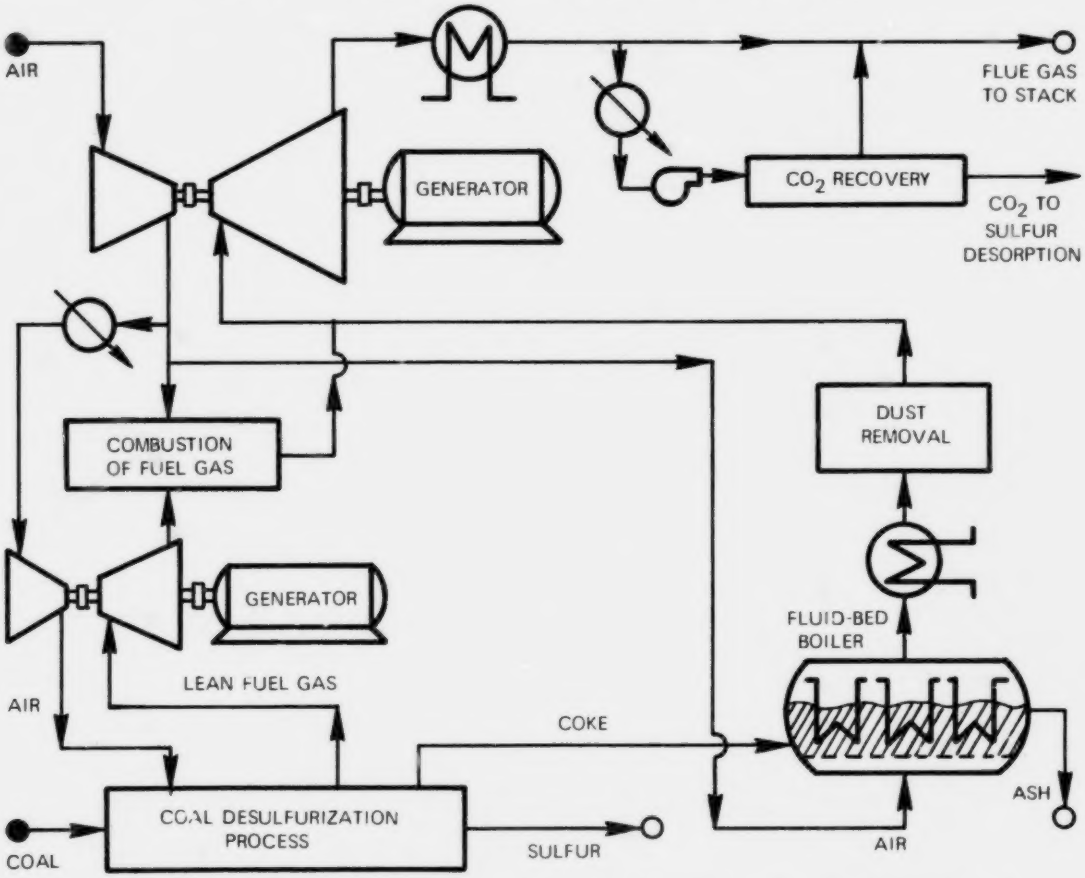
AIR
GENERATOR
FLUE GAS TO STACK
CO2 RECOVERY
CO2 TO SULFUR DESORPTION
COMBUSTION OF FUEL GAS
DUST REMOVAL
GENERATOR
FLUID-BED BOILER
LEAN FUEL GAS
AIR
COKE
ASH
COAL DESULFURIZATION PROCESS
COAL
SULFUR
AIR

Figure 2. Lean fuel gas from coal desulfurization used to raise the temperature of combustion products from a coke-fed pressurized fluid-bed boiler.

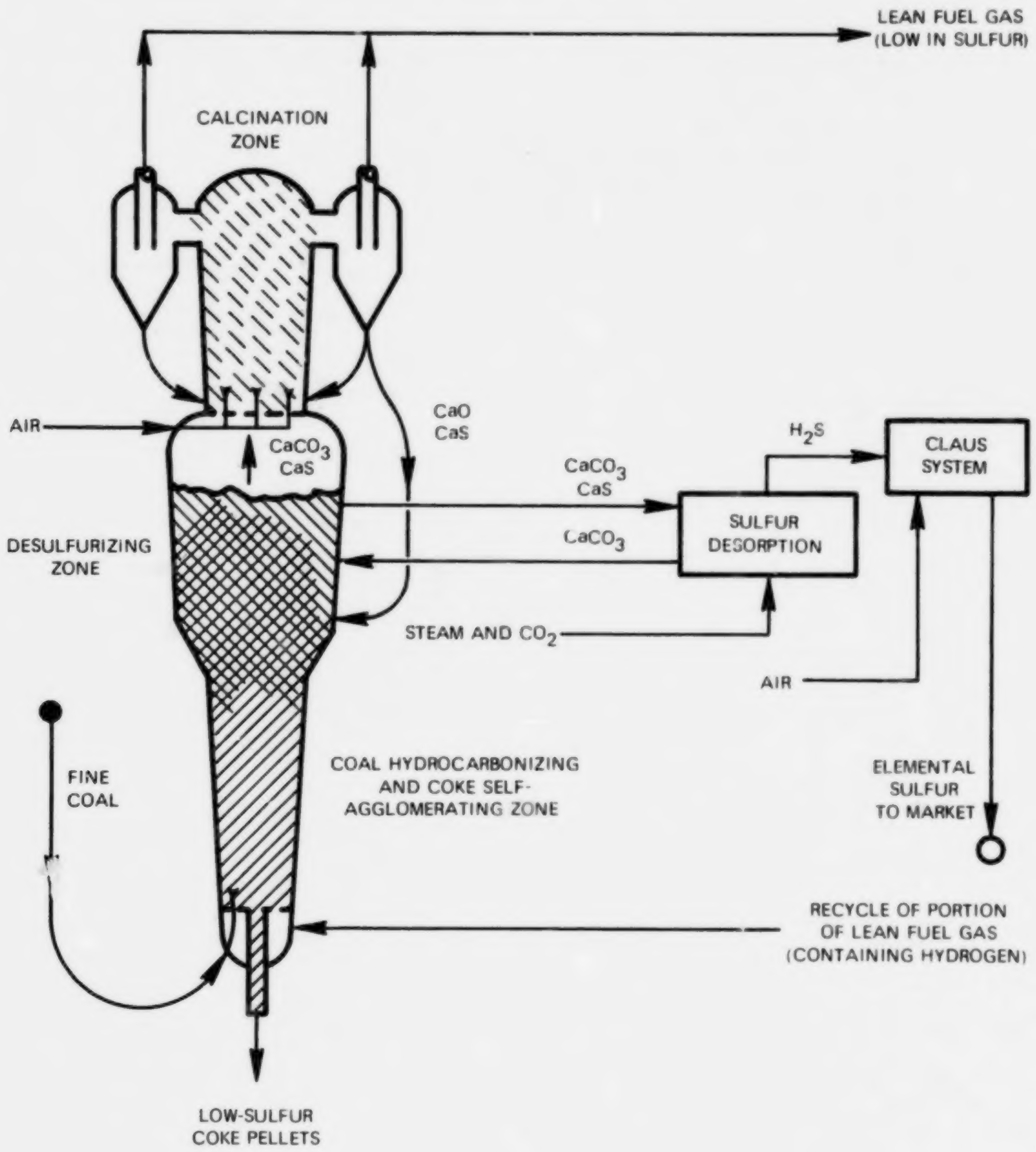
LEAN FUEL GAS (LOW IN SULFUR)
CALCINATION ZONE
AIR
CaO
CaS
$CaCO_3$
CaS
H_2S
CLAUS SYSTEM
$CaCO_3$
CaS
SULFUR DESORPTION
$CaCO_3$
DESULFURIZING ZONE
STEAM AND CO_2
AIR
FINE COAL
COAL HYDROCARBONIZING AND COKE SELF-AGGLOMERATING ZONE
ELEMENTAL SULFUR TO MARKET
RECYCLE OF PORTION OF LEAN FUEL GAS (CONTAINING HYDROGEN)
LOW-SULFUR COKE PELLETS

Figure 3. Acceptor process converting coal into low-sulfur fuels.

2. FLUIDISED-BED COMBUSTION AND THE DESIGN OF BOILERS

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INTRODUCTION

Conventional electricity-generation boilers, whether fired by pulverised coal or oil, are characterised by very large combustion chambers. The size is dictated: first, by the need to allow sufficient time for combustion despite temperatures in the range 1200-1600°C; and secondly, by the need to allow molten and potentially corrosive ash constitutents to resolidify before coming into contact with the banks of convective superheater tubing. The result is a supporting structure more than 200 ft high, for a 660-MW boiler, costing some 14 percent of the total boiler cost with, in addition, extensive civil works to accommodate the substantial point loads.

Fluidised beds have two properties which allow a radical rethinking of boiler design concepts.

1. The rapid movement of particles within the fluidised bed gives rise to relatively high rates of heat and mass transfer between the solids and the fluidising gas. The result is that coal can be burned in a fluidised bed of some 2-3 ft deep at volumetric heat release rates in excess of anything achievable by conventional methods and at mean bed temperatures between 800 and 850°C. At these relatively low temperatures: ash constituents do not melt; almost all the alkali metals, known corrosion promoters, are retained in the bed; and, by the addition of limestone to the bed, a substantial proportion of the sulphur content of the fuel can be retained as $CaSO_4$. Thus the need for a large combustion volume is eliminated.

2. The rapid movement of the particles within the fluidised bed gives rise to relatively high heat-transfer coefficients between the bed and surfaces immersed in it, up to 5 times greater than those achievable in conventional gas-tube heat exchangers. The environment of a fluidised-combustion bed allows steam-raising tubes to be placed in the bed. Thus, substantial savings can be made in the length of tubing required to extract a given amount of heat, and possibly, the less corrosive environment will allow less expensive alloys to be used in the superheater and reheater sections.

DESIGN OF FLUIDISED-BED BOILERS

The most important decision in the design of a fluidised-bed boiler operating at atmospheric pressure is the choice of fluidising velocity. The fluidising velocity fixes the oxygen availability, and hence heat-release rate, per unit cross-sectional area of bed which, in turn, fixes the total cross-sectional area required for a given steam duty. Therefore, considered in isolation, the capital cost of the boiler falls with increasing fluidising velocity because of savings in containment costs.

However, two factors oppose the incentive to increase fluidising velocity.

1. As the fluidising velocity is increased, a progressively coarser size spectrum of bed particles must be used in order to limit elutriation losses. As the mean particle size of a fluidised bed increases, the heat-transfer coefficient within it decreases: e.g., as the mean particle size increases from 0.4 mm (0.016 in.) to 1.0 mm (0.039 in.), the coefficient falls about 35 percent. The result is a tendency for the bed depth, and hence pressure drop and running costs, to increase in order to accommodate the increased area of tubing required in the bed; this is in addition to the increased capital cost of tubing.

2. As the fluidising velocity increases, elutriation rates, and hence carbon loss from the bed, tend to increase due to the increased agitation and degradation within the bed, and residence time in the combustion zone tends to decrease. Therefore, larger and more elaborate recycle systems are required to achieve any particular combustion efficiency with a consequent increase in both capital and running costs.

Thus the choice of fluidising velocity becomes an optimisation of conflicting economic incentives.

If, on the other hand, the boiler is pressurised, an additional degree of freedom is introduced in that, for a given velocity and a given cross-sectional area, the heat release may be increased by increasing the pressure, and hence oxygen availability. Therefore, for a given steam output the containment cost can be reduced by increasing the pressure. There is no heat transfer penalty because velocity, and hence bed size spectrum, remains constant; however, bed height, and hence pressure drop, increases because more heat must be extracted from a bed of fixed cross-sectional area, and there is a limit to the density of tube packing. The apparent insensitivity towards operating under pressure is, however, offset by the increased plant complexity imposed by the pressure and also by the availability of industrial gas turbines designed for a suitable pressure ratio and capable of being modified without incurring substantial development charges. Although the remainder of this discussion will consider only atmospheric pressure boilers, much of it will be relevant to the case of operation under pressure.

Having chosen a fluidising velocity, the cross-sectional area of the unit is fixed for a given steam output. Because the bed temperature will be limited to between 800 and 900°C by ignition and ash fusion criteria, respectively (at least for English coals), the required distribution of heat-transfer surface between the fluidised bed and conventional surfaces above the bed may be calculated. Typically some 60 percent of the heat release must be extracted directly from the bed if a temperature of about 850°C is to be maintained.

There is a cost incentive to place those parts of the steam/water circuit where the more exotic materials have to be used in the fluidised bed. If carbon steel has a cost index of 1.0 (under English conditions), 2 percent Cr has 2.4, 9 percent Cr has 5.4, and Austenitic 316 has 13.4. Therefore, the incentive is to put the superheater and reheater in the fluidised bed and make up any deficiency with evaporator surface leaving the water walls and any gas-space surface to the remaining vaporisation and the economiser. However, under part-load conditions, the downsteam ends of in-bed reheaters and superheaters can get relatively hot, requiring the use of austenitic steels where ferritic steels are adequate at the continuous-rated output. Thus, particularly at relatively low fluidising velocities with consequently high heat-transfer coefficients, there may be an economic incentive to place the downstream sections of the reheater and superheater in the gas-space above the bed and avoid the use of expensive steels.

Finally, there is an incentive (at least under English conditions) to maximise factory construction, thereby minimising site work and the consequent scope for construction errors—all of which reduce the time of construction and hence the interest charges incurred. Interest during construction can be as much as 26 percent of the capital cost of a 2000-MW power station. Fluidised-bed combustion presents, for the first time, the possibility of designing a large boiler as a number of semi-independent units which can be factory-assembled, leaving the minimum of connecting-up on site. For English road transport conditions, the maximum transportable size is about 14 ft wide x 40 ft long x 16 ft high; however, for a coastal power station, sizes as large as 25 x 44 x 22.5 ft can be brought in by sea. As an example, using road transport criteria, a 120-MW boiler operating at a fluidising velocity of 8 ft/s can be designed as five factory-assembled beds (two evaporator, two reheat, and a superheat) with a secondary superheater and an ecomoniser in the gas-space above the bed. This arrangement is estimated to save 9 months in construction time. With high interest charges, the incentive to utilise factory assembly can be strong enough to decide a fluidising velocity at the expense of say a slightly lower combustion efficiency.

CONCLUSION

The foregoing shows that many of the potential advantages of fluidised-bed combustion, for reducing the capital cost of boiler plants, are self defeating unless the design is optimised in respect to each variable.

The design should:

1. Obtain maximum heat-transfer coefficients in the bed and the bulk of the boiler surface operating at metal temperatures in excess of 400°C immersed in the bed.

2. Obtain the maximum combustion efficiency by maximising the residence time of gas and particles in the bed and the combustion space above it.

3. Obtain maximum factory assembly in packaged units.

4. Reduce structural and civil work to a minimum.

5. Reduce to a minimum the lengths of high-pressure pipework connections.

Design aims 1 and 2 argue for low fluidising velocities; aims 3, 4, and 5 argue for high fluidising velocities. Thus, if full advantage is to be taken of the potential of fluidised-bed combustion for reducing the cost of power station plant, an economic optimum must be sought whereby the fullest advantage is taken of all possible savings.

ACKNOWLEDGMENTS

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3. IGNIFLUID BOILERS FOR AN ELECTRIC UTILITY

R. H. DEMMY

UGI Corporation

INTRODUCTION*

The UGI Corporation operates an electric utility (Hunlock) in Northeastern Pennsylvania, serving an area of 440 square miles near Wilkes-Barre, along the Susquehanna River. UGI's Hunlock Generating Plant (88,700-kw net) burns anthracite from the Northern and Middle Pennsylvania anthracite fields on Barley stokers and in pulverized-fired boilers. This plant's latest addition, completed in 1959, is a Foster Wheeler 400,000 lb/hr pulverized boiler utilizing anthracite bank material with a quality of approximately 9000 Btu/lb. Stack emission is controlled by electrostatic and mechanical precipitators installed by Research-Cottrell.

UGI's Planning Department indicated that new generation was required in the early 1970's, both to meet new sales and to replace old facilities. To satisfy this projected need for new generation, we investigated the possibility of burning fuels adjacent to our service territory. We found that Babcock-Atlantique of France had a boiler utilizing the Ignifluid combustion process which would be capable of burning the anthracite culm banks of our area (there are 100,000,000 tons within 40 miles of our projected plant site).

Small samples of the culm bank material were sent to France: first, four 5-lb bags; later 40 tons in 55-lb bags. When preliminary laboratory tests indicated that a full-scale combustion test should be attempted, approximately 800 tons of 40-percent ash, 7500 - 8000 Btu/lb culm bank fuel was shipped to an area in the French Alps near Grenoble, where the Cartonneries de la Rochette operates a cardboard factory.

OBSERVATIONS OF PRODUCTION TESTS

Today, I would like to discuss with you our observations in both Casablanca, Morocco, where the largest Ignifluid boiler is located, and in La Rochette, France, where the combustion tests were performed.

In Casablanca

Arriving in Casablanca late in the afternoon, we were concerned that we would be unable to visit the plant that day. We proceeded immediately to the power plant, approximately 1 mile from the center of Casablanca.

As we approached, we were shocked to see that the stack indicated no power generation. Because we were approaching an anthracite plant we had anticipated what we have experienced in the United States in the way of anthracite stacks; seeing the stack completely without any visible emission, we were quite upset that we had travelled such a distance to find the plant not operating.

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*The technical assistance of P.A.Mulcey, fuel consultant of Dallas, Pennsylvania, is acknowledged in the development of this paper.

When we arrived in the control room, the Moroccan operators told us that the plant was at full load, a fact which was difficult for us to accept. But the recording charts indicated that the plant, with a maximum capacity of 60 mw, was operating at its normal capacity, approximately 55 mw, and that the boiler had been operating at approximately the same output most of the day.

The stack emission control for this boiler is a Lurgi electrostatic precipitator preceded by a mechanical precipitator. The precipitator was to have been a three-cell unit but the Moroccans, without sufficient money to buy all three cells, purchased two internals and the shell for the third. Results were so successful that they have not added the third electrostatic section.

The dust removal from these units is similar to what we are used to in the States; a hydrovac or similar system is used. The one difference is that all of the dust which is collected by the precipitators is reinjected into the furnace. There is no fly ash pond. All ash from the Ignifluid boiler is removed as bottom ash, and sluiced into the Atlantic Ocean which, at the site of the power plant, is 400-ft deep.

The burnout of the carbon is quite complete: 4 to 6 percent carbon is left in the ash.

Electrostatic precipitation is successful because the fly ash of an Ignifluid boiler is high in carbon and therefore low in resistivity which is the opposite of the pulverized boilers operated in the States. Therefore, a clean stack is not a desire but a fact. For an operator of an anthracite-fired plant or a bituminous coal low-sulfur plant which would again have high resistivity, it was indeed hard for us to believe our own eyes when we saw a boiler at full load with an optically clear stack.

In La Rochette

Visiting the site of our production tests in the French Alps at La Rochette, we found that the Ignifluid boiler there has no electrostatic preci- pitators, but two mechanical precipitators. The stack emission at that site was similar to the best emission we have at Hunlock Plant, utilizing electrostatic precipitators installed last year.

La Rochette's coal is transported by rail from Antwerp, Belgium, and dumped directly into the silos at La Rochette.

The developer of the Ignifluid process (described more fully at the end of this paper) is Ms. Albert Godel, of Société Anonyme Activit, who spoke at the First International Conference in 1968, and is still very active and interested in this process. Babcock-Atlantique has purchased the patents from him and are directing the development program.

A steam flow of 100,000 lb/hr from a grate area approximately 3 ft wide by 25 ft long (compared to 100,000 lb/hr from a standard anthracite spreader stoker 20 ft wide by 28 ft long) indicates the improvement in the state-of-the-art of burning anthracite.

The tests were under the supervision of CERCHAR, the French equivalent of the U.S. Bureau of Mines and Bureau of Standards, combined.

The continuous removal of the ash from the Ignifluid boiler is accomplished by an inclined grate rising at an angle of 10-12 degrees. The level is maintained in the fire bed by having variable air pressure supplied to the various zones of the fire by separate air ducts.

In this boiler there are six zones; the highest pressure is where the raw fuel is supplied to the boiler by a Redler feeder. The pressure decreases as the fuel progresses toward the front of the furnace; the area where the ash rises out of the fluidized bed requires additional air for final combustion of the carbon in the ash.

During our test, the combustion was stopped due to a plugging of the coal feeder and we were able to witness a reignition of boiler. The coal is supplied to the small grate in a manner similar to the way a spreader stoker would receive its coal; the fire is started in the same manner, with excess air supplied until the fire starts effervescing. When this effervescent condition is more or less uniform over the entire grate, which is not moving at the time, the bed is then fluidized by the introduction of sufficient air to cause the particles to rise from the grate and be balanced by the upward flow of the primary air and the action of gravity on the particles.

Air distribution in the boiler does not seem to be critical. In this boiler, ambient air (approximately 50 percent primary air and 50 percent secondary air) is introduced just above the fluidized bed in the same zone as is the reinjected fly ash. When the bed is fluidized, the furnace looks like a gas furnace with an incandescent white heat.

An inspection of the internals of this boiler indicated the tubes to have no slag accumulation. The 2-year-old grate looked brand new, undoubtedly because of the introduction of primary air to keep the grate from reaching high temperatures. The ash carbon burnout for our tests, as was predicted in model tests in the laboratory, was not as high as those in Casablanca (4-6 percent) and La Rochette (6-8 percent): the carbon in the ash of the Pennsylvania culm was 17-20 percent. In the final design of the proposed boiler, burnout to a level of less than 10 percent will be accomplished by a post-combustion furnace similar to the type utilized for expanded ash for lightweight aggregate purposes.

PROPOSED NANTICOKE STATION

After evaluating the combustion tests in France and completing the overall engineering evaluation, it was determined that a 300,000-kw station could be built utilizing two Ignifluid boilers and one turbine-generator. A unit size of 300,000 kw was selected due to the availability of units of this size already operating. To further enhance the availability, two boilers are being used. Since the major cause of loss of generation is boiler failure, the use of two boilers will allow operation at half load while repairing the failed boiler. Steam conditions at the throttle of the turbine will be 1800 psig 1000°F/1000°F. The turbine will also be supplied with the option for extraction: adjacent to the power plant site, the Commonwealth of Pennsylvania is planning to install a pilot plant for acid mine-water demineralization which will require approximately 300,000 lb/hr of low-pressure steam.

The fuel for this plant is above ground and available. No deep mining of coal will be required for the proposed 30-year life of the plant. The fuel will come from existing anthracite culm banks (refuse from the preparation of anthracite), 100,000,000 tons of which are located within 40 miles of the plant site. This fuel will be upgraded to approximately 8000 Btu/lb, a heating value which the boiler has proven itself capable of handling. Material not taken to the power plant will be left at the bank sites and regraded into usable contours so that all of the present areas which are not usable as real estate today will be available for real estate development. The high ash content (40 percent) of the fuel would normally cause a considerable problem in handling; however, this is not anticipated as the ash has proven itself to be excellent for utilization in cinder blocks and has potential use as a lightweight building aggregate.

We believe that this plant will meet the ecological requirements of today's society.

1. We do not anticipate particulate emission problems.
2. Since the fuel has less than 0.5% sulfur, there will be no SO_2 emission.
3. NO_x emission will be reduced. Tests in France showed that the NO_x emission of an Ignifluid boiler was approximately half that of a pulverized boiler firing the same coal.
4. Thermal discharge to the Susquehanna River will be controlled by the utilization of cooling towers.

THE IGNIFLUID PROCESS APPLIED TO UTILITY BOILERS*

The Ignifluid process is a fluidized-bed method of combustion of small coal which is maintained in stable aerodynamic suspension under the effect of an ascending air current which gives the layer the appearance of an incadescent boiling liquid, hence the trademark Ignifluid.

The process utilizes a standardized fluidized combustion burner at the base of the boiler. Consisting of a very narrow inclined stoker of less than one-tenth of the total section of the boiler, it supports the fuel bed which is fluidized by a controlled primary air current; it also extracts the clinker from the bed in its ascending rearward motion.

Secondary air, blown over the top of the fluidized bed, completes combustion. All the grit carried over is cycled back on the surface of the fluidized bed, with the result that all the ash from the coal is extracted as clinker by the chain-grate.

A high rate of combustion is obtained (over 300 lb of coal per sq ft) because combustion takes place in the whole 2-ft-deep mass of the turbulent bed and not only on the surface of the stoker, as is the case with conventional stokers or spreaders.

An Ignifluid-fired boiler burns any type of small fuel: sized from 1/8 in.-0 to 3/4 in.-0, from anthracite to bituminous coal, coking or not, and regardless of the ash content. The efficiency is as high as with pulverized firing.

Mechanically, the Ignifluid unit is similar to the chain or traveling-grate stoker, including as it does the same drive and return sprockets, traveling grate or chain, and wind-box arrangement consisting of six or more compartments.

Essential Differences

The essential differences are:

1. The grate width is only about 10 percent that of the traditional anthracite stoker.
2. The grate is tilted 8-12 degrees from the horizontal, in the direction of travel of the grate, as contrasted to the level stoker.
3. There is a different under grate air-pressure pattern from front to back—substantially a complete reversal—with the maximum (main duct) pressures about 50 percent higher.
4. The coal feed is different: instead of being carried by the stoker from a feed hopper into the furnace under a leveling gate, coal is dropped by a Redler conveyor into a steep-angled pipe which intersects the furnace front wall a few feet above the grate. Air injected through the bottom of the feed pipe tends to spread the coal outward and forward as it falls toward the fluidized bed which in turn, has an inherent spreading proclivity due to the boiling action.

On a stoker, the wind-box pressures usually range drom 1/2-in. water in the first compartment, to a maximum of 4-5 in. in the fourth, and grading off to 3 in. in the sixth. In the

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* The assistance of F.W. Kuehn, engineering consultant of Allentown, Pennsylvania, is acknowledged for this portion of the paper.

Ignifluid unit, pressure is highest (about 8 in. of water) in the first compartment; it grades down uniformly to 1 in. in the sixth, where the coal bed is fluidized in normal operation, as distinguished from start up (which will be explained).

As might be surmised, with a chain width only 10 percent that of the stoker, the burning or reaction is much more intense.

Startup

To understand the action, let us run through a startup. A small amount of coal is fed into the unit so it lies on the grate to a depth of 6-12 in. at the front. The induced draft fan is started. Next, oil or gas torches are lighted and directed down onto the surface of the coal until it is uniformly ignited. The forced draft fan is started and undergrate (called primary air) air pressure is slowly built up in the compartments as indicated, with a straight-line sloping pattern from maximum pressure in the first (or front) compartment to a minimum in the last.

As the flow and pressures are increased, a point is reached where the coal lifts off the grate and suspension firing begins. At this point, the fluidity action is rather unstable, and the air flow must be increased further at constant pressures to somewhere under the point in velocity where most of the particles would have left the fluidized bed (become gas borne). Fortunately, because there is a wide band of velocities possible between the low or unstable point at which the bed tends to collapse, and the high point where the bed would be carried off with the gas, the operation is inherently very stable.

Under normal operation then (from full load down to about 15 percent of rating), the top surface of the fluidized bed is level, looking like boiling lava. Depth above grate ranges from a maximum of about 20 in. at the front (over the first air compartment) to nothing but the clinker thickness over the fifth air compartment of a six-compartment unit.

When the coal particles are all consumed, the remaining bits of fine ash become sticky and agglomerate with others forming dense clinker which falls on to the grate and is carried over the rear sprocket in a ribbon 1-3 in. deep.

Blankets of dead coal cover and protect the sloping refractory walls along the sides and front of furnace, below the water-cooled walls. The dead coal layers are maintained because there is no air flow in these areas to promote and support combustion.

Vertical water jackets, extending from the top grate line down to the refractory at base of the crickets outside the wind boxes, form a barrier between the moving hot clinker on the grate and the dead coal; they also support the latter.

An air seal, at the interface between the bottom of structural bridge across the furnace and the top of the moving chain grate just ahead of the first undergrate air compartment, prevents any loss of siftings in this area, especially during startup.

Very little loss of fuel in the form of siftings is experienced through the grate, it is claimed, because of the higher undergrate air pressures used (as compared to conventional stokers) and to the small air openings. Individual stoker chain links are notched in the middle to facilitate separation of the clinker at the rear sprockets.

Secondary air, which in larger units is usually preheated to higher temperatures than the primary air, is introduced into the furnace through side walls a few feet above the fluidized bed to complete combustion of the fuel rich gases.

An inclined air nozzle under the rear arch is used to blast any fine coal particles which drop onto the surface of the clinker bed back into the active combustion zone.

Control

Control is essentially conventional, and typical of later automatic stoker control in the States except for a unique fuel-bed thickness probe and controller. Primary air is controlled by deviation of a master steam-pressure controller from a set point, steam pressure being sensitive to load changes. As primary air flow increases or decreases under direction of the master controller, it changes the rate of coal combustion and, therefore, the depth of the fluidized bed. A water-cooled fuel-bed thickness probe, inserted in the bed just above the grate near the front of the furnace, monitors the thickness of the bed, and passes its signal continuously to a fuel-bed thickness controller. The controller, set to hold the fluidized bed at a constant thickness, passes on the deviation-from-normal signal to the fuel-bed controller. Secondary air flow is trimmed by desired-percentage CO_2 in furnace exit gases. Judging from typical air duct sizing, secondary air appears to be about 50 percent of the total air. Induced-draft fan dampers, in turn, hold a constant furnace draft of minus 5 mm.

Operating Advantages

Operating advantages claimed for the Ignifluid unit over the stoker, which taken together account for its ability to efficiently burn low-grade coals containing up to 50 percent ash, include:

1. Automatic spreading of coal inherent.
2. Clinkers sink through fluidized bed to grate.
3. Intimate and intense reaction of air and fuel.
4. Fluidized bed completely pervious to air. (With a bed in a fluid state, it is obvious that the cleanliness of the coal is not important; in fact, the coal can be unwashed.)
5. Loss of siftings through air holes in grate chain link is not as great as in conventional stoker firing because of higher air pressures required by the process which in turn means smaller air holes through the grate. (Claimed riddling or sifting losses are as low as 0.5 percent.)

Unusually high thermal efficiencies (upwards of 88 percent) are claimed, even without air preheating when using a medium volatile (6 percent), reasonably well prepared (16.4 percent ash), fairly dry (6.6 percent moisture) anthracite of proper sizing (zero to 5 mm). Boiler efficiency of 81.74 percent was guaranteed for the UGI units.

As the coal becomes poorer (i.e., higher in ash), a significant higher loss to be reckoned with is that in the sensible and latent heat loss in the clinker, part of which will be reclaimed with a post-combustion furnace.

Forced outage rate for the 55-metric ton evaporation per hour Ignifluid burning unit at La Rochette (given to Société Babcock & Wilcox by the owner-operator paper company, Cartonneries de La Rochette) for the year August, 1966 to August, 1967 was 2.14 percent: 0.3 percent was due to the stoker alone; 1.84 percent was for such things as repairs to ash-reinjection system, coal-handling system breakdowns, and replacement of belts on the forced draft fan.

CONCLUSION

The ecological demands of the United States today are forcing electric utilities not to consider coal-fired plants due to the severe problems of electrostatic precipitation of high resistivity ash which results from either pulverized-anthracite or low-sulfur-bituminous firing. With insufficient natural gas available, the utilities are leaning toward low sulfur oil and nuclear material as the future sources of energy for electrical generation. If the coal industry is to participate in the electric utility energy market, a modification in the method of burning fuel is indicated. We believe that the fluidized bed as proposed by Babcock-Atlantique is the only method of fuel combustion available today in sizes which can be upgraded to the large steam production requirements of the electric utility industry of the United States. Although this method is available in commercial size today, we fear that the delays inherent in developing commercial pressurized fluidized beds will prohibit their commercial development in the United States. However, if the Ignifluid process is introduced into the United States, we will have fuel technologists knowledgable in fluidized combustion capable of guiding the development of the next step forward in this technology; namely, pressurized fluidized beds.

4. THE MODULAR FLUIDIZED-BED BOILER CONCEPT

J. W. BISHOP

Pope, Evans and Robbins

ABSTRACT

The development of the Pope, Evans and Robbins modular fluidized-bed boiler concept is described. The early and unsuccessful attempts to design a fluidized-bed boiler along more "conventional" lines are also described and the lessons drawn from these failures presented. A preliminary conceptual design and cost estimate is presented for an electric utility boiler having a capacity of 300,000 pounds of steam per hour.

INTRODUCTION

The purpose of this paper is to describe the experimental and design history leading to the presently proposed modular design concept of a fluidized-bed boiler, along with some details of the most recent concept of a 300,000 lb/hr small utility unit.

EARLY HISTORY

In 1965, as part of our program to extend the capacity of packaged coal-fired boilers, the late LeRoy F. Deming visited various European combustion and boiler developers, including BCURA. It was obvious that fluidized-bed firing and direct transfer of heat from the bed to the heat transfer surface represented the most promising system for providing a high-capacity coal-burning boiler.

Imbedded-Tube Concept

The first effort was to develop a boiler concept as a guide for the ensuing experimental work. Using BCURA's initial project of 1,000,000 BTU/hr/ft^2 of bed area, the boiler concept of Figure 1 was developed. This concept consisted of a continuous imbedded-tube bank unit with a longitudinal steam drum and three lower headers. Coal was to have been screw-fed at two locations in the hope that the fluidizing air issuing from the grid would lift and distribute the coal.

The atmospheric steam boiler of Figure 2 was then built, originally with a longitudinal screw-feeding mechanism and a perforated plate grid. Of course, the air did not distribute properly and the coal merely caked above the screw.

Water-Cooled Column

We then digressed to a water-cooled column wherein the air distribution problem was solved: first, with a "rock sandwich" grid (i.e., a layer of rock between two perforated plates); later, with the "button grid" currently in use. This system directs air down onto the grid plate, precludes dead spots, prevents sifting into the plenum, and permits operation at over 2000°F, a condition which has not been approached with other grid plate designs. Relying on BCURA's experience, we screw-fed the coal into the column with air jets blowing the coal off the screw, but we were unable to duplicate the BCURA operation. We then developed a system for dropping coal into a pneumatic transport line.

Coal-Distribution Problems

The boiler of Figure 2 was placed in operation, but problems of coal distribution were encountered in the presence of the imbedded tube bank. Any agglomeration of particles between tubes rapidly formed clinkers which stuck between the tubes. Although we could remove these clinkers, by lifting the boiler off the grid plate plenum assembly, it was obvious that this could not be done with an actual boiler. Erosion was heavy on the lower bank of tubes. The lessons learned here were:

1. Avoid use of imbedded tube banks if possible.
2. If imbedded tube banks are required, consider the effect on fuel distribution, access for cleaning, and the minimum height above the distributor to preclude erosion. (Design for heat removal is only one of a number of considerations.)

The imbedded tube bank system was abandoned.

The design of the atmospheric-pressure boiler had been predicated on a heat transfer coefficient between the bed and the imbedded tubes of 50 Btu/hr/ft^2/°F. It was found, however, that this coefficient was not useful in reproducing either the furnace exit gas temperature or the bed temperature. It was necessary to almost double the value of the measured coefficient to reproduce the performance with a given imbedded surface area. A sizable fraction of the released energy was removed from the bed by mechanisms other than direct contact heat transfer. It was necessary (as in the case of the later BCURA shell-and-tube boiler experience) to cut and remove tubes so as to reduce the heat removal rate so that, in turn, a deeper bed could be operated in which good combustion could be achieved. The lessons learned were:

1. A measured heat transfer coefficient is useful in predicting the heat flux into a single tube.
2. Bed temperature and exit gas temperature cannot be predicated on the basis of wetted surface area and a heat transfer coefficient in a water-cooled enclosure. An empirical correlation has to be derived as a prediction tool.

Open Modular Concept

Because so little "wetted" surface was needed to control bed temperature if "viewed" surface were available, the design concept of Figure 1 was seen to be unworkable, and the problems inherent in that design unnecessary. The simplest way to avoid coal distribution and maintenance problems was to go to the open modular concept of Figure 3.

By selecting the distance between tube walls, the required heat exchange surface (wetted and viewed) could be provided, still retaining a relatively large maintainable open combustion space. If fuel and air to each module were controllable, it would be possible to achieve a larger turndown.

The first checkout of this system was performed on the boiler of Figure 2 by halving the width (to 18 in.) and removing the tubes on one side. Pending construction of an actual single-module unit, the modified boiler of Figure 2 was operated to develop coal feeders and light-off procedures.

Coal Feed and Flue Gas Travel

The system which provided the most even coal distribution was an up-through-the-grid method. However, with single-screened coal, any moisture resulted in plugging of the feed tube. Also, erosion at the turns was excessive.

Therefore, a straight-in coal feed system was optimized and adopted for use on the single-module boiler (FBM). Excessive carryover of bed material was noted in all FBX test runs. This was attributed to the gases taking a horizontal path above the bed, entraining the material without the allowance for particle recovery provided by a straight vertical express gas path. This assumption was verified in later FBM tests: a 6-percent ash coal held the bed level; i.e., made up for attrition elutriation losses. The lessons learned included:

1. Do not specially prepare the coal used in the development work; use coal identical to that to be used in practice.
2. Do not consistently feed as-received coal pneumatically if an upward directed turn is required, at least not where forced draft air is the motivating force. (Compressed air has not been used because of economics.)
3. Avoid horizontal movement of the flue gas until it has passed through a screen such as a convection bank, superheater, or tubular air heater.
4. Bed particles will carry over if gases take a horizontal turn even at heights well above the theoretical disengaging height.
5. A vertical express route for flue gas provides for simple trouble-free design.

Initial FBM Test Results

The FBM design (see Figure 4), based on the lessons learned in the FBX, utilized a direct coal injection system and incorporated an integral system for fly ash reinjection. Both coal and fly ash were fed to the same boiler bed.

Figure 5 graphically presents a heat balance during typical operation. Note the high loss from unburned carbon in the fly ash. Utilizing, in a separate operation, a smaller fluidized-bed column (the FBC), insulated so as to reduce heat loss to the walls, collected fly ash was refired, and the carbon loss was reduced to an acceptable level. See Figure 6.

The result of this experiment was the Carbon-Burnup Cell—a separate combustion zone or segregated region for which the primary fuel is fly ash.

During the initial test phase, the present system of light-off was developed. By "puddling" a flame into one section of the bed surface, ignition is accomplished, followed by propagation of the fire to all portions of the module through openings in the dividing tube wall.

It is realized that acceptable carbon burnup has also been achieved in a single fly ash reinjected combustion space by utilizing extensive uncooled refractory surfaces above the bed and imbedded heat exchange surface area, utilizing the same principle as the refractory arch in a conventional anthracite-fired boiler. In other words, if the elutriating unburned carbon leaving the bed can be held long enough in a hot non-radiant heat-removing zone, the fly ash carbon will burn out. A column employing 80 percent of its total height and volume as a refractory-lined, non-heat-transferring, elutriated-carbon burnup section has demonstrated acceptable carbon loss. The combustion of carbon in this extensive freeboard area results in a temperature rise in that area, further enhancing the carbon combustion potential. However, such a system can only be applied to boiler design employing a large open "furnace" above the bed, thus defeating the objective of compact combustion and lowered cost

In addition to the above size and cost consideration, the integral (one combustion zone) concept requires multiple recycle of fly ash with a consequent increase in dust loading to the collectors. For this reason, present particulate emission standards are currently forcing existing pulverized coal and spreader stoker fired units to restrict recycle of fly ash. The once-through separate carbon-burnup cell precludes the "multiplication" of dust loadings.

Early Modular Boiler Concepts

In addition to the initial concept of Figure 3, other preliminary designs based on initial experimental work were prepared, and performance predicted for efficiencies ranging from 85 to 93 percent. These early concepts have included:

1. A 250,000-pound-per hour packaged boiler designed for the production of 250-psig saturated steam.
2. A 250,000 pound-per-hour packaged boiler designed for the production of 600-psig, 750°F steam
3. A 2,000,000 pound-per-hour boiler designed for the production of 2000-psig. 1000°F steam (and for reheat to 1000°F).
4. An 8,000,000 pound-per-hour boiler designed for the production of 2400-psig. 1000°F steam (and for reheat to 1000°F).

Further Development in the FBM

One of the important design features that required checking was the effect of bed communication between adjacent modules, particularly between a boiler cell operating at a relatively low temperature and a carbon-burnup cell (CBC) operating at a high temperature.

The FBM was modified and a small insulated region was added. (See Figure 7.) While this small column is not a realistic CBC, which would be much larger, it was the right size for the quantity of fly ash generated by the FBM.

To ensure that good communication and equal bed height were obtained, an open area of 90 square inches was initially provided between the FBM and the CBC.

Initial operation of the FMB/CBC was poor. With the FBM at 1600°F, the CBC could not reach 2000°F, the temperature required for adequate burnup. The major cause was the rapid interchange between beds through the 90-square-inch opening. When there is too much intercommunication between the two zones, the desired temperature difference cannot be maintained. Reduction of the opening of 2 square inches was found to be adequate for light-off and bed-level equalization. This small opening permitted the desired temperature to be achieved.

A second energy loss resulted from the carryover of hot bed material into the horizontal duct. It was also impossible to determine burnup performance since the collected ash was heavily contaminated with bed material.

This carryover problem was solved by the addition of a BCURA-conceived baffle screen. However, the screen acts as a remarkably effective heat sink. A high (approximately 40 Btu/ft^2 hr °F) heat transfer coefficient has been computed for this device.

A vertical coal feeder has been tested. The uniformity of coal distribution can be determined by measuring oxygen content at various points above the bed. Oxygen in flue gas may be held to within ±0.2 percent across the width and length of the bed with coal feeders as developed in this work. The technique lends itself to a simple coal delivery system. Figure 8 shows the present FBM boiler setup in the Alexandria, Virginia, laboratory.

The lessons learned from this work include:

1. Very small (but properly located) openings between regions permit equal bed levels and interbed light-off.
2. Tubular baffle screens, properly positioned, serve as effective heat exchangers, almost as good as "wetted" surface, and may also be used in lieu of a high freeboard.
3. Downward-directed vertical pneumatic coal feeders provide for an even fuel distribution over a relatively large area

and greatly simplify the problem of boiler coal supply.

4. Cold air, supplied by a forced draft fan, can provide all the motive force required to inject coal into a fluidized bed.

Carbon loss from the FBM/CBC, when employing a water-cooled baffle screen directly above the bed, is as high as that found in a typical spreader stoker operation (see Figure 9). The present CBC is simply too short* and the baffle screen is too effective as a heat exchanger. By replacing the water-circulated baffle with uncooled alloy steel rods, satisfactory combustible loss (0.8 percent, see Figure 10) has been attained. Laying out and equipping the unit with total freedom could have provided a design for a thermal efficiency in the 90-percent range. Although the present FBM/CBC will not achieve an overall thermal efficiency much greater than 80 percent, much has been learned from its operation; contemplated revisions do not include a departure from the basic modular concept.

PRESENT DESIGN CONCEPTS

Present design concepts are:

1. An open fluidized-bed combustion space should be utilized so as to minimize coal distribution problems and to provide adequate access for boiler maintenance.

2. Coal injectors should be served with motive air supplied directly from a forced-draft fan.

3. Heat transfer surface should be proportioned between direct-contact, viewed surface, and baffle surface so as to transfer directly from the bed 50-60 percent of the energy released in burning the fuel.

4. Flue gas should follow a long vertical express route, including convection and economizer banks.

5. The design of the boiler should permit all boiler cells to communicate with the CBC, especially if a regenerative SO_2-capture method is to be used.

Based on these concepts, a 300,000-lb/hr shippable unit has been conceived for an eastern utility. (See Figures 11 and 12.) The modules (cells) run parallel to the steam drum and connect to a single CBC. At least until additional experimental work can be done on coal-feed techniques, a 4.5-ft spacing for coal feed points seems advisable. The design shows eight fuel-receiving points, each serving two adjacent locations. Coal and limestone feed (where used for SO_2 abatement) are combined. Primary superheaters are shown in the bed, but may also be arranged to serve as baffle screens above the bed. When a module is shut down, superheating stops, providing an automatic attemperation mechanism. Some external attemperation will still be required. Fly ash from the boiler cells is collected in the primary cyclone and fed to the CBC via parallel circuits to four mushroom feeders. There is also provision for auxiliary coal addition. No point in the CBC bed is farther than 2 feet from a feed point. Control of the CBC is based on maintaining constant bed temperature. The CBC is cooled by the secondary superheater, taking advantage of the higher available temperature differentials in the section. Preliminary predicted data for this unit is outlined in Table 1.

Referring to Figure 11, it should be noted that two of the vertical bed-immersed tubes extend into the bed, completely surrounded by the bed particles. As suggested by Seibel of Erie City, a few rows of tubes (say 2 or 3) could extend from the walls, forming a restricted tube bank, enhancing heat transfer but not interfering with the dispersion of coal feed or the maintenance accessibility of the boiler. This would permit widening the cells (reducing the required number of cells

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* The CBC, added 2 years after the FBM was installed, had to be located beneath the steam drum. The distance between grid and horizontal discharge is only 40 in.

Table 1. PRELIMINARY DESIGN DATA FOR 300,000-POUND-PER-HOUR FLUIDIZED-BED UTILITY BOILER (1500-PSIG DESIGN, 1270-PSIG OPERATING, 925°F)

Service	Duty, MBtu	ΔT diagram, °F	Tubing data Surface ft^2	Tubing data Length, ft (O.D., in.)
Final superheater in CBC (In-bed)	11.2	2000↔2000 860 → 925	223	430 (2)
(Above-bed)	5.4	2000 →1270 860← 834	203	390 (2)
Primary superheater in boiler cells	64.0	1600↔1600 575 → 834	1,640	3,130 (3)
Evaporation (In-bed)	98.4	1600↔1600 575↔575	1,930	2,460 (3)
(Particle recovery screen)	74.9	1600↔1600 575↔ 575	1,820	1,460 (3) 1,280 (2)
(Convection)	25.1	1600 →1270 575↔ 575	2,980	5,700 (2)
Economizer	48.0	1270 → 709 525← 385	9,480	18,100 (2)
Regenerative air heater	35.4	709 → 274 70 → 550	-	-
Totals	362.4	--------	18,276	29,030 (2) 3,920 (3)

Bed data	Boiler section	CBC section
Grid area, ft^2	364	45
Air rate, lb/hr ft^2 : superficial velocity, fps	794:11.7	730:12.8

Fan data	Grid	Bed	Convection & economizer	Air heater	Total
Draft loss, in. w.g.	5	16	5	5	31

Fan power @ 76,560 SCFM, 31 in. w.g. = 498 HP
Heat balance data, MBtu: Fuel input = 358.6; stack loss 16.6;
unburned combustible = 7.2; radiation and misc. = 7.4; boiler efficiency = 91.8%.

for a given capacity) and/or permit the reduction of bed level (and draft loss). Arrangements incorporating this concept are under study and will be incorporated in the design.

A preliminary cost estimate has been prepared based on post-development conditions but using 1970 costs. This estimate, summarized in Table 2, shows a basic boiler cost

Table 2. PRELIMINARY COST ESTIMATE FOR 300,000-LB/HR FLUIDIZED-BED UTILITY BOILER

1. Basic boiler		
a) Steam drum	106,500 lbs	
b) Drum headers	6,400 "	
c) Drum internals	4,100 "	
d) Tubes	51,600 "	
e) Lower headers	28,800 "	
f) Casing	3,000 "	
f) Contingency	59,600 "	
Total weight	260,000 lbs	
Estimated cost @ $1.32/lb	$343,200	
Add 50% for special construction	171,600	
Total basic boiler		$514,800
2. Boiler appurtenances		
a) 409 ft^2 of air distributor	$ 40,000	
b) Economizer (30,000 lb)	30,000	
c) Air heater (120,000 lb)	55,000	
d) Boiler trim	25,000	
e) Coal/ash/additive supply	200,000	
f) Ducts/plenum/breeching	40,000	
g) Insulation (30,000 lb)	60,000	
h) F.D. fan and drive	40,000	
i) Collectors (2)	30,000	
j) Electrostatic precipitator	290,000	
k) Controls and instrumentation	120,000	
Total boiler appurtenances		930,000
3. Normal design; installation and contingency		
a) Installation (includes miscellaneous piping and electrical	350,000	
b) Contingency	200,000	
c) Normal design	120,000	
Total design, installation, and contingency		670,000
Total normal installation at existing plant		2,114,800
Add 25% for first prototype unit		528,700
Estimated prototype facility cost		$2,643,500

of $514,800 and a total plant cost of $2,114,800. For the first installation, we have applied the multiplier of 1.25 to the estimated cost for post-development conditions. The cost for the first unit is, therefore, estimated at $2,643,500.

CONCLUSIONS

During the past several years, a number of design arrangements have been proposed for fluidized-bed boilers. Three true boilers have actually been built. two by Pope, Evans and Robbins; followed by BCURA's shell-and-tube unit. PER's "FBM" is the only unit currently in service. In two cases it was found necessary to remove some or all of the imbedded tubes, since at the time these two designs were completed, the removal of energy from the bed by mechanisms other than direct contact heat transfer was not anticipated. All three of these actual boilers have exhibited unacceptable carbon losses. The use of a segregated fly ash carbon-recovery section operating at high temperature (2000° F) has been demonstrated as a means of recovering the unburned carbon without resorting to low velocities and excessively large bed areas (pressurized fluidized-beds excepted).

The use of extensive closely spaced steam generating tube banks immersed in the bed (particularly horizontal banks requiring forced circulation) are felt to be an unnecessary complication for most fluidized-bed boilers. The arrangement of tubes in single-row banks so as to form walls has been demonstrated in the FBM to be a useful design concept. The modular approach is the only concept that has thus far been extensively tested under actual operating conditions.

In summary, it is felt that the design concepts proposed here offer the following advantages:

1. Maximum flexibility.
2. Simplest coal and sulfur acceptor feed capability.
3. Maximum accessibility for maintenance.
4. Good turndown capability.
5. No requirement for an induced draft fan.
6. The use of natural, rather than forced, circulation.
7. Compact construction by the use of high-mass gas flows and heat release rates.
8. Low bed particle carryover; ability to operate with low-ash coal.
9. Segregated fly ash combustion region; capable of operation at optimum burnup conditions, without compounding the particulate emission problem. (This feature also obviates the need for large furnace-like freeboards for the consumption of unburned carbon.)
10. Capability of using a "built-in" limestone regeneration process.
11. Low capital costs.

Regardless of the direction taken by fluidized-bed boiler development, it is felt that experience should be effectively disseminated and considered. There can be no good reason for reintroducing design features already shown to be deficient–unless a new "twist" has been conceived to obviate the previously demonstrated deficiency. Boiler design features must be based upon data derived from large-scale experimental equipment duplicating, or very closely simulating, the proposed arrangement.

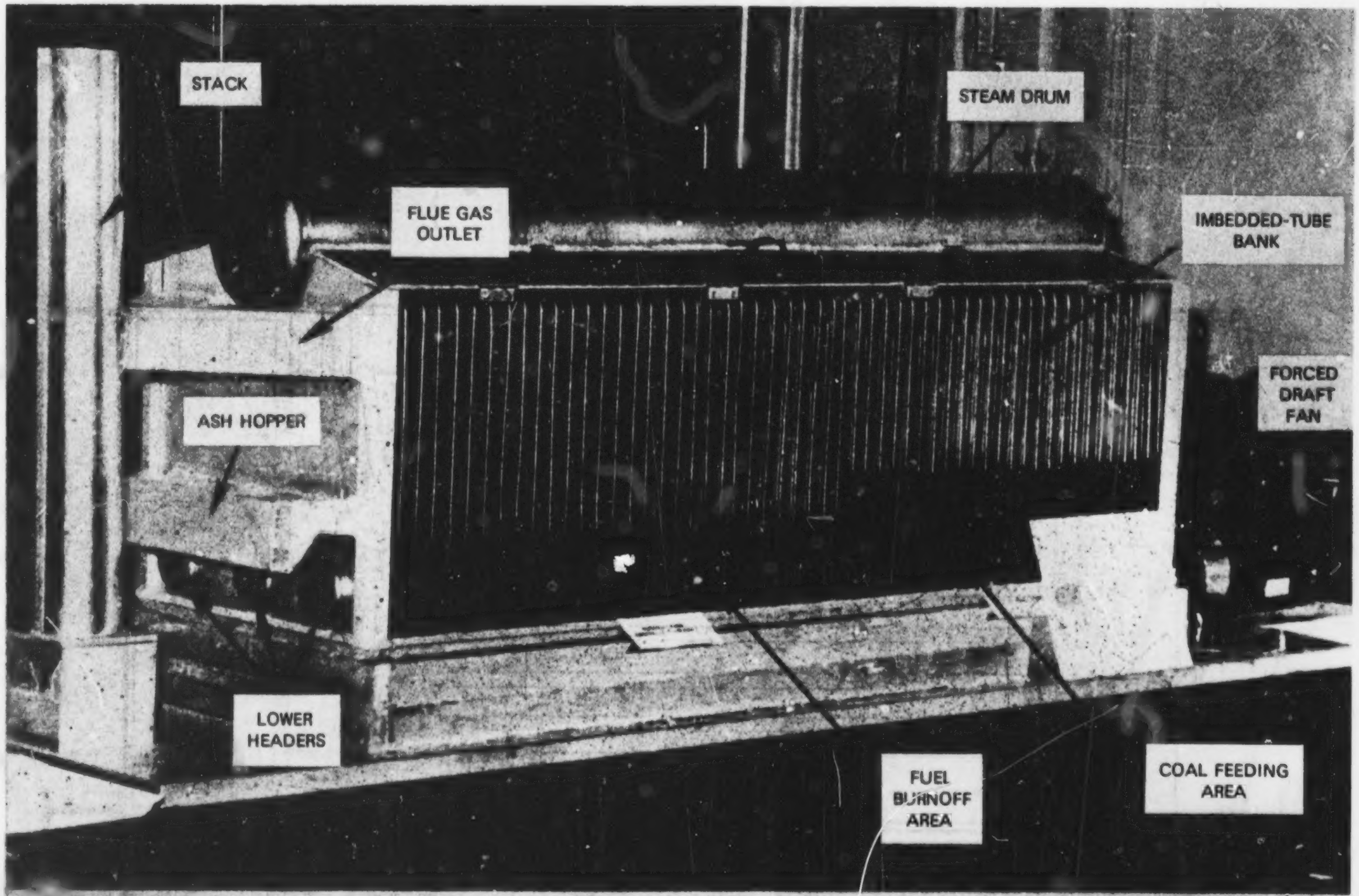

STACK
STEAM DRUM
FLUE GAS OUTLET
IMBEDDED-TUBE BANK
ASH HOPPER
FORCED DRAFT FAN
LOWER HEADERS
FUEL BURNOFF AREA
COAL FEEDING AREA

Figure 1. Plastic model, packaged fluidized-bed boiler.

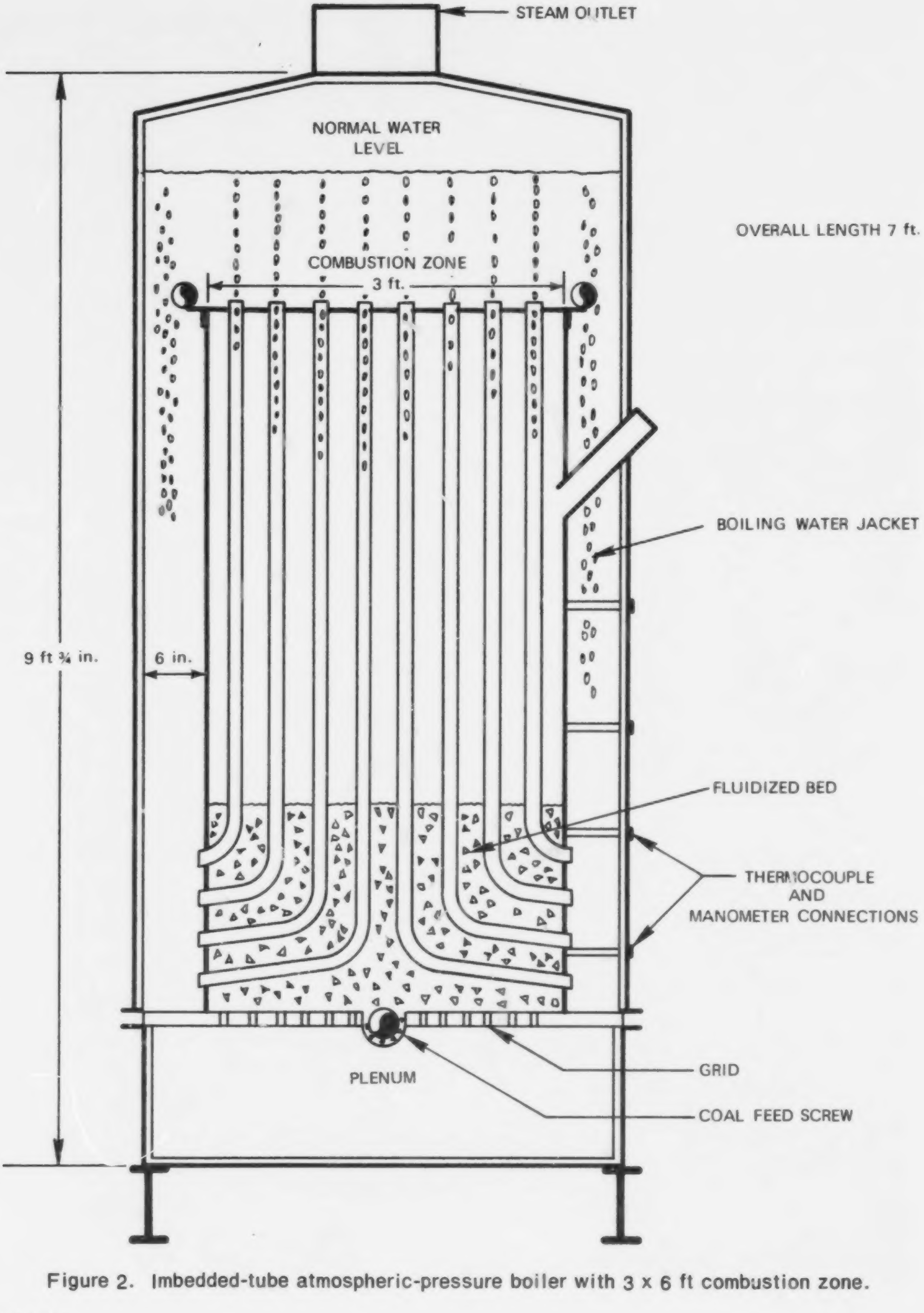

STEAM OUTLET
NORMAL WATER LEVEL
OVERALL LENGTH 7 ft.
COMBUSTION ZONE
3 ft.
BOILING WATER JACKET
9 ft ¾ in.
6 in.
FLUIDIZED BED
THERMOCOUPLE AND MANOMETER CONNECTIONS
GRID
PLENUM
COAL FEED SCREW

Figure 2. Imbedded-tube atmospheric-pressure boiler with 3 x 6 ft combustion zone.

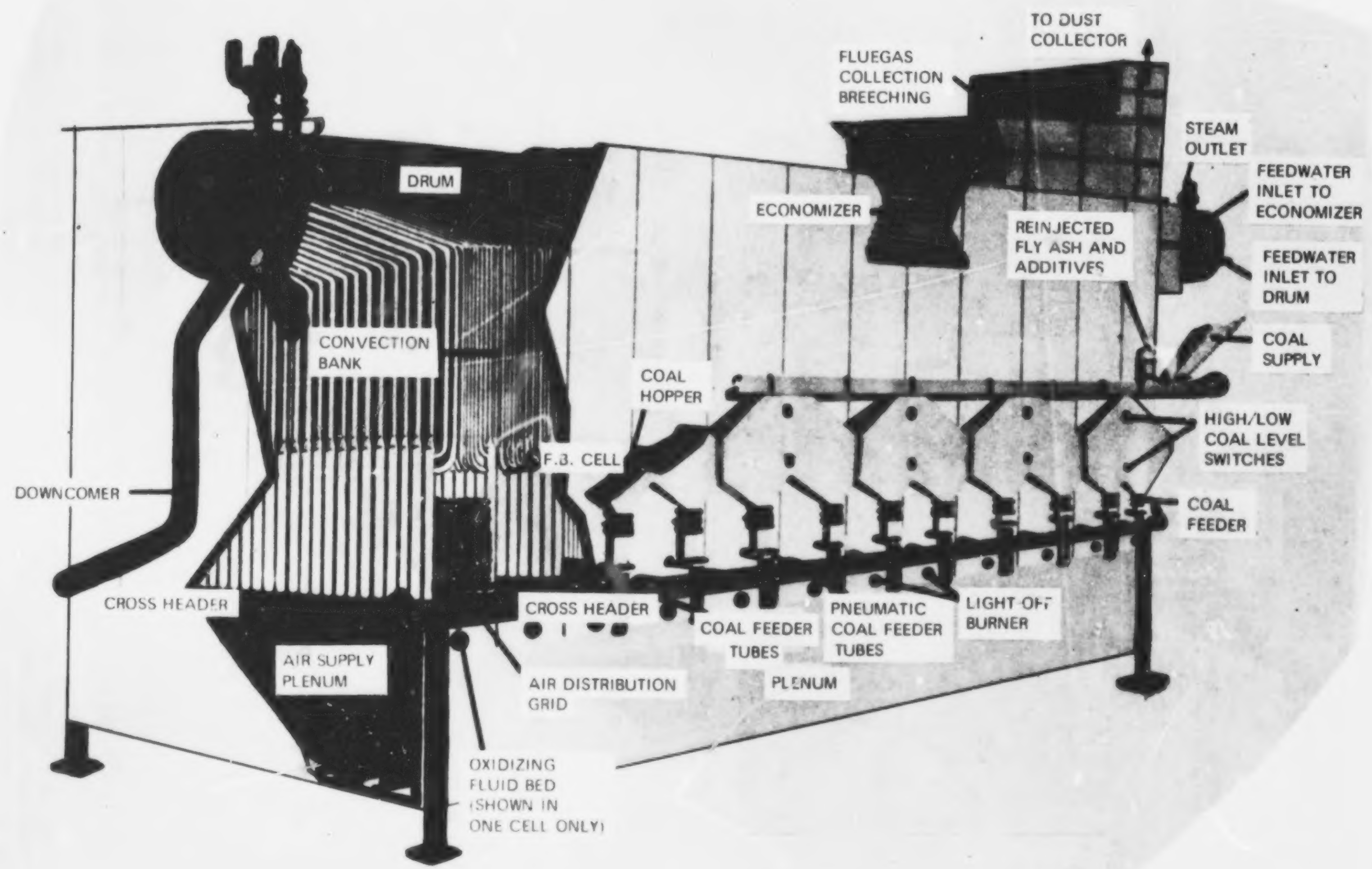

TO DUST COLLECTOR
FLUEGAS COLLECTION BREECHING
STEAM OUTLET
DRUM
FEEDWATER INLET TO ECONOMIZER
ECONOMIZER
REINJECTED FLY ASH AND ADDITIVES
FEEDWATER INLET TO DRUM
CONVECTION BANK
COAL SUPPLY
COAL HOPPER
HIGH/LOW COAL LEVEL SWITCHES
F.B. CELL
DOWNCOMER
COAL FEEDER
CROSS HEADER
CROSS HEADER
LIGHT-OFF BURNER
PNEUMATIC COAL FEEDER TUBES
COAL FEEDER TUBES
AIR SUPPLY PLENUM
PLENUM
AIR DISTRIBUTION GRID
OXIDIZING FLUID BED (SHOWN IN ONE CELL ONLY)

Figure 3. Industrial modular boiler (original concept).

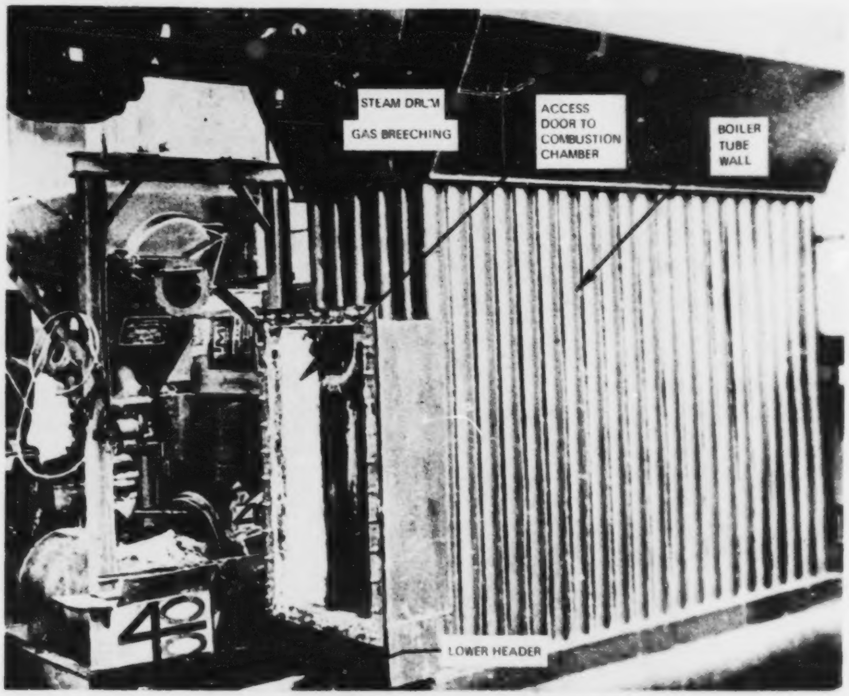

STEAM DRUM
GAS BREECHING
ACCESS DOOR TO COMBUSTION CHAMBER
BOILER TUBE WALL
LOWER HEADER

Figure 4. Fluidized-bed boiler module (FBM).

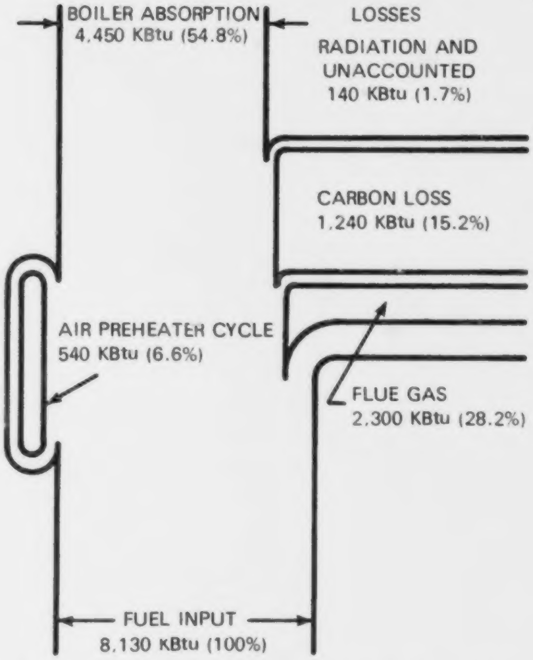

BOILER ABSORPTION
4,450 KBtu (54.8%)
LOSSES
RADIATION AND UNACCOUNTED
140 KBtu (1.7%)
CARBON LOSS
1,240 KBtu (15.2%)
AIR PREHEATER CYCLE
540 KBtu (6.6%)
FLUE GAS
2,300 KBtu (28.2%)
FUEL INPUT
8,130 KBtu (100%)

Figure 5. FBM heat balance without carbon recovery (boiler convection and economizer heat transfer deleted).

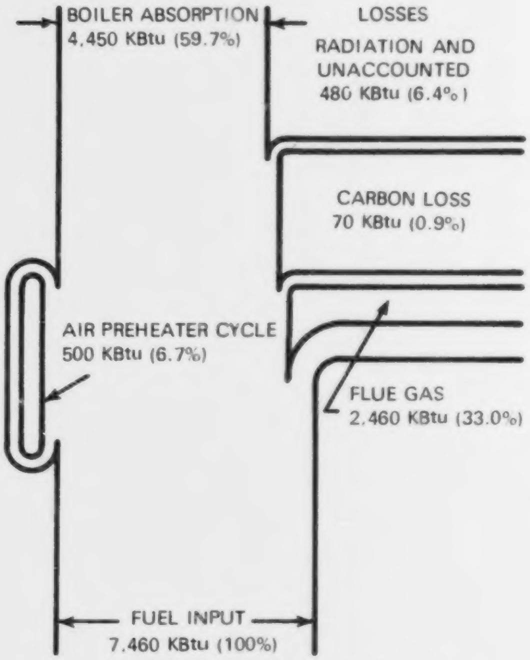

BOILER ABSORPTION
4,450 KBtu (59.7%)
LOSSES
RADIATION AND UNACCOUNTED
480 KBtu (6.4%)
CARBON LOSS
70 KBtu (0.9%)
AIR PREHEATER CYCLE
500 KBtu (6.7%)
FLUE GAS
2,460 KBtu (33.0%)
FUEL INPUT
7,460 KBtu (100%)

Figure 6. FBM heat balance with carbon recovery (boiler convection and economizer heat transfer deleted).

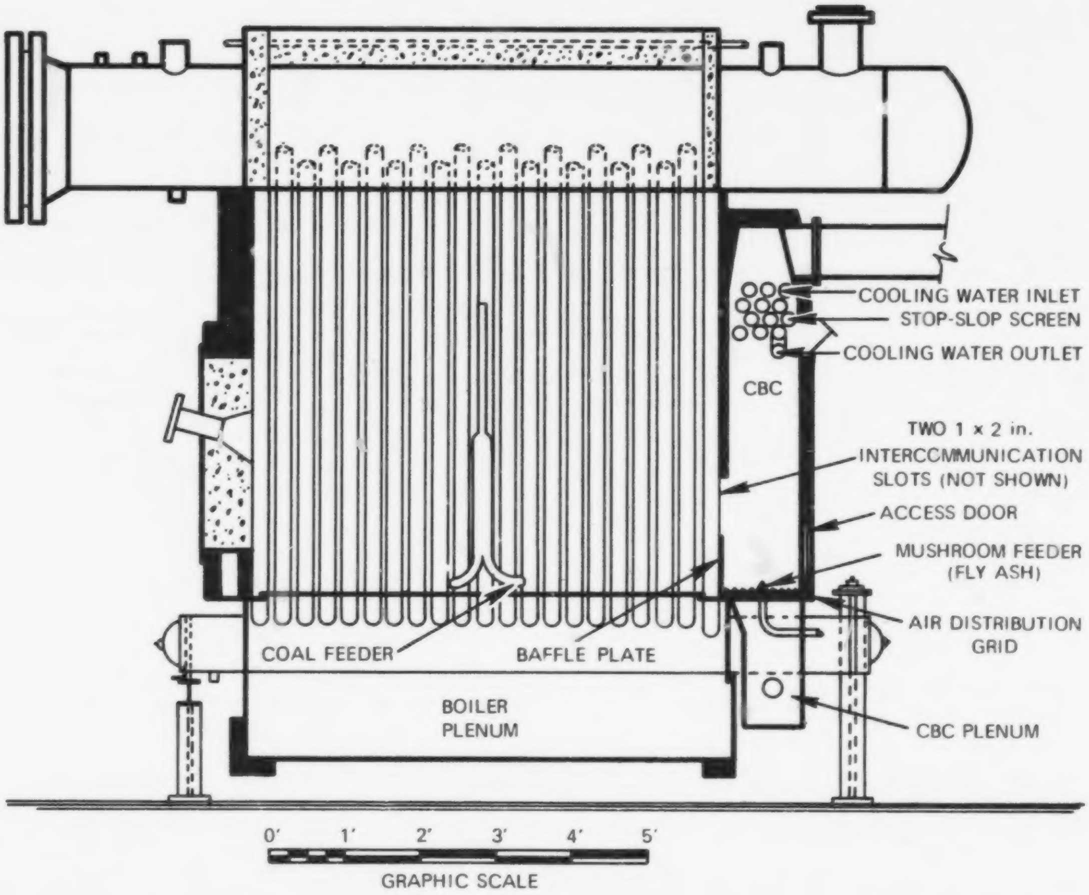

COOLING WATER INLET
STOP-SLOP SCREEN
COOLING WATER OUTLET
CBC
TWO 1 x 2 in. INTERCOMMUNICATION SLOTS (NOT SHOWN)
ACCESS DOOR
MUSHROOM FEEDER (FLY ASH)
AIR DISTRIBUTION GRID
CBC PLENUM
COAL FEEDER
BAFFLE PLATE
BOILER PLENUM
0'
1'
2'
3'
4'
5'
GRAPHIC SCALE

Figure 7. Present boiler arrangement.

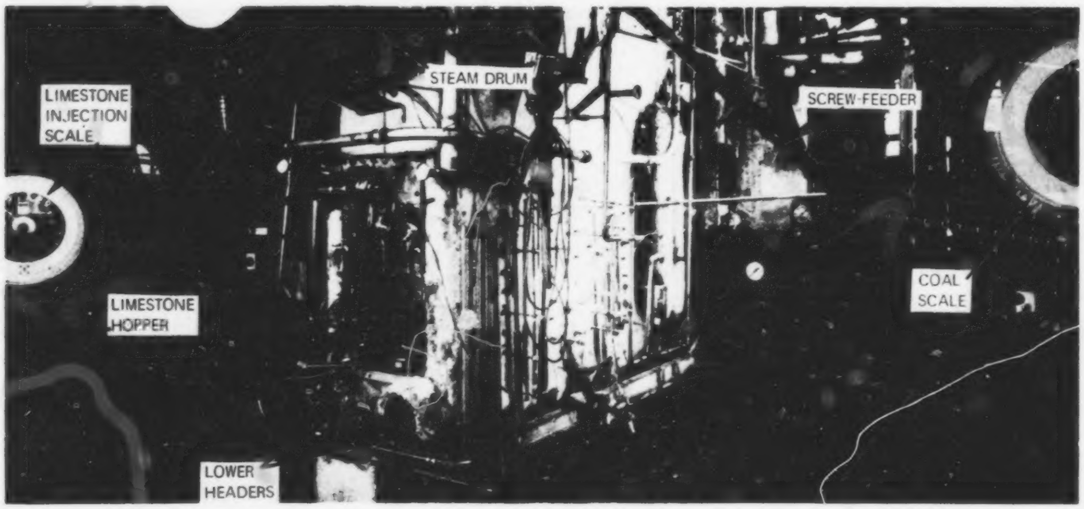

LIMESTONE INJECTION SCALE
STEAM DRUM
SCREW-FEEDER
LIMESTONE HOPPER
COAL SCALE
LOWER HEADERS

Figure 8. Test boiler, Pope, Evans and Robbins Alexandria, Virginia, laboratory.

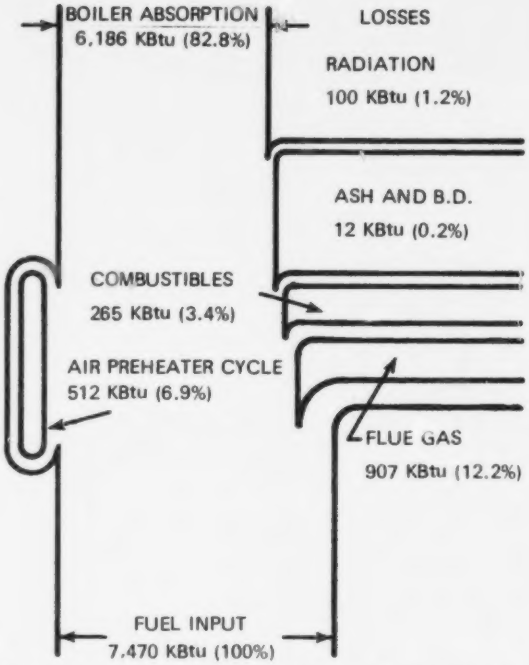

BOILER ABSORPTION
6,186 KBtu (82.8%)
LOSSES
RADIATION
100 KBtu (1.2%)
ASH AND B.D.
12 KBtu (0.2%)
COMBUSTIBLES
265 KBtu (3.4%)
AIR PREHEATER CYCLE
512 KBtu (6.9%)
FLUE GAS
907 KBtu (12.2%)
FUEL INPUT
7,470 KBtu (100%)

Figure 9. FBM test B-5 heat balance (with integral CBC water-cooled stop-slop screen).

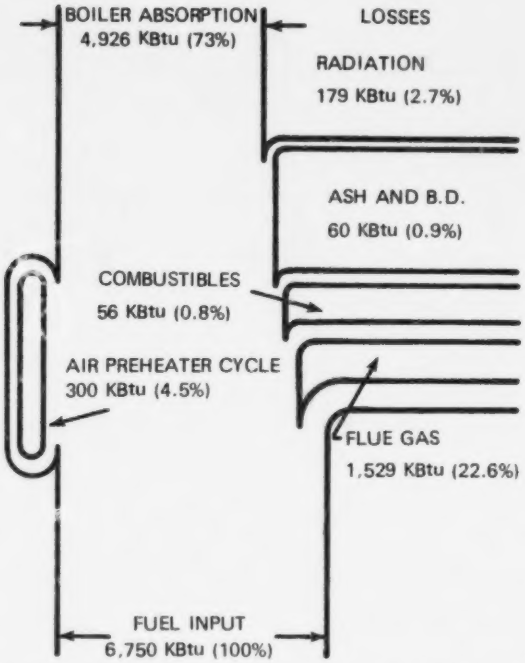

BOILER ABSORPTION
4,926 KBtu (73%)
LOSSES
RADIATION
179 KBtu (2.7%)
ASH AND B.D.
60 KBtu (0.9%)
COMBUSTIBLES
56 KBtu (0.8%)
AIR PREHEATER CYCLE
300 KBtu (4.5%)
FLUE GAS
1,529 KBtu (22.6%)
FUEL INPUT
6,750 KBtu (100%)

Figure 10. FBM test B-14 heat balance (with integral CBC, uncooled stop-slop screen).

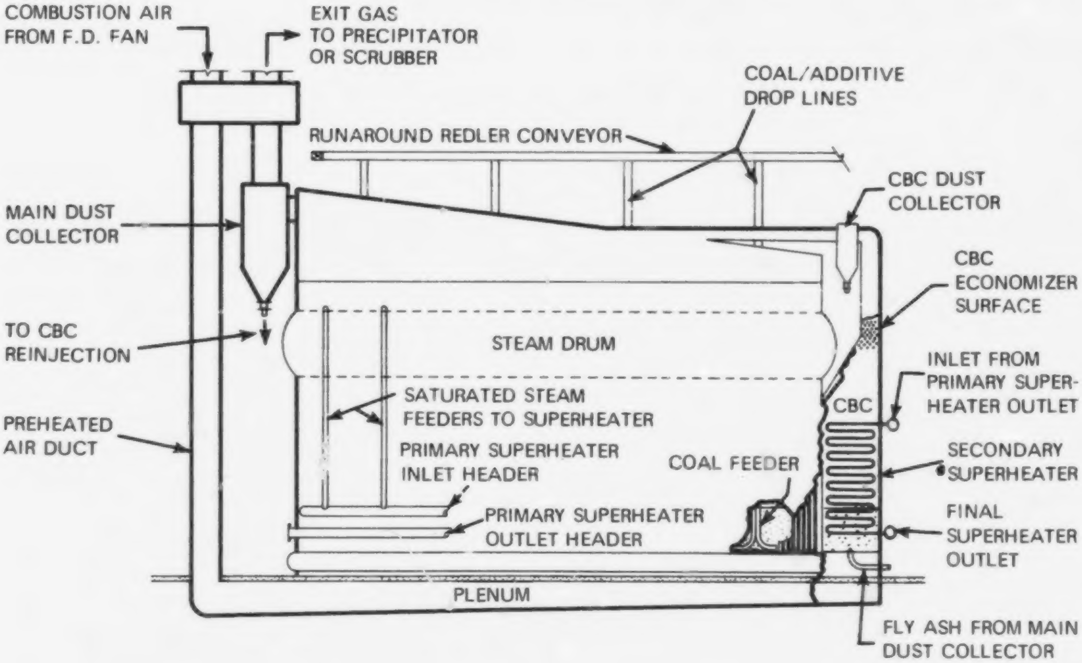
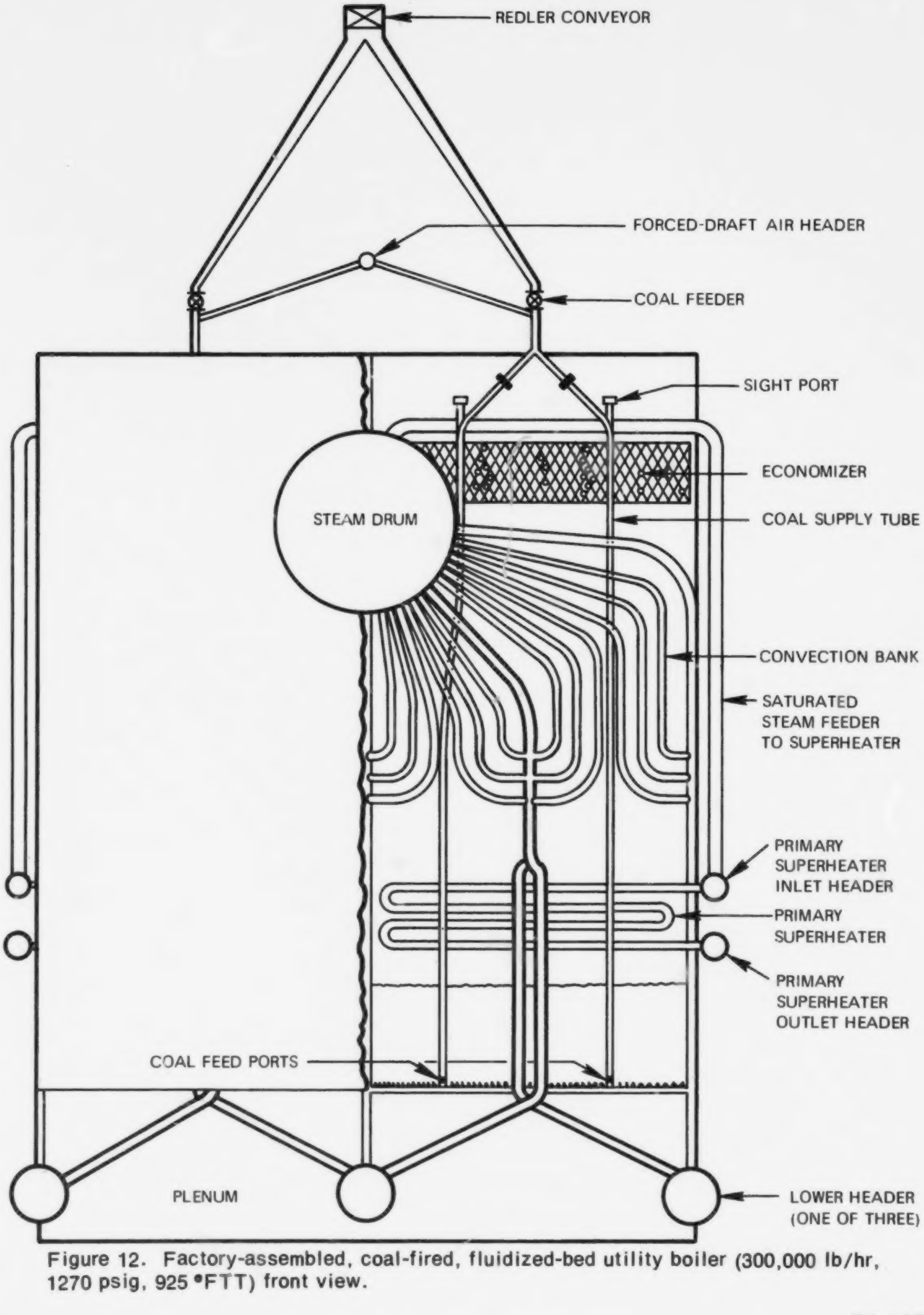

COMBUSTION AIR
FROM F.D. FAN
EXIT GAS
TO PRECIPITATOR
OR SCRUBBER
COAL/ADDITIVE
DROP LINES
RUNAROUND REDLER CONVEYOR
CBC DUST
COLLECTOR
MAIN DUST
COLLECTOR
CBC
ECONOMIZER
SURFACE
TO CBC
REINJECTION
STEAM DRUM
INLET FROM
PRIMARY SUPER-
HEATER OUTLET
CBC
SATURATED STEAM
FEEDERS TO SUPERHEATER
PREHEATED
AIR DUCT
PRIMARY SUPERHEATER
INLET HEADER
COAL FEEDER
SECONDARY
SUPERHEATER
PRIMARY SUPERHEATER
OUTLET HEADER
FINAL
SUPERHEATER
OUTLET
PLENUM
FLY ASH FROM MAIN
DUST COLLECTOR

Figure 11. Factory-assembled, coal-fired, fluidized-bed utility boiler (300,000 lb/hr, 1270 psig, 925 °FTT) side view.



REDLER CONVEYOR
FORCED-DRAFT AIR HEADER
COAL FEEDER
SIGHT PORT
ECONOMIZER
STEAM DRUM
COAL SUPPLY TUBE
CONVECTION BANK
SATURATED STEAM FEEDER TO SUPERHEATER
PRIMARY SUPERHEATER INLET HEADER
PRIMARY SUPERHEATER
PRIMARY SUPERHEATER OUTLET HEADER
COAL FEED PORTS
PLENUM
LOWER HEADER (ONE OF THREE)

Figure 12. Factory-assembled, coal-fired, fluidized-bed utility boiler (300,000 lb/hr, 1270 psig, 925 °FTT) front view.

5. FLUIDIZED-BED BOILERS—CONCEPTS AND COMPARISONS

D. L. KEAIRNS AND D. H. ARCHER

Westinghouse Reserach Laboratories

INTRODUCTION

Fluidized-bed boilers offer economic and social benefits of compact, cheap, and efficient power generation systems; clean air; and effective use of natural resources and raw materials for fuel. Westinghouse, as part of a contract with the National Air Pollution Control Administration, is preparing and evaluating boiler designs in order to provide a technical and economic measure of the potential of fluidized-bed combustion boilers for air pollution control. These concepts must then be compared between themselves and with conventional boiler or power systems.

Both new and existing concepts are being considered. Fluidized-bed boiler concepts have been generated by considering various fluidized-bed configurations and operating conditions. Operating conditions are selected based on the evaluation of fluidized-bed combustion data, fluidized-bed technology as applied to other processes, and boiler and power system requirements. The critical fluidized-bed combustor parameters are calculated from heat and material balances, heat transfer relations, kinetics of combustion and desulfurization, and fluid flow relations given the configuration, operating conditions, pollution control limitations, and customer specifications. These parameters include cross-sectional area of the bed, heat transfer surface requirements, bed height, bed composition, pressure drop through the system, solids and gas distribution, and auxiliary equipment specifications.

The projected investment and operating cost and pollution control capabilities of the fluidized-bed boiler power systems considered can be compared with each other and with other power systems. The best power system is the one which meets customer specifications and pollution control limitations at the minimum cost.

A pressurized boiler power system is used to illustrate a fluidized-bed concept and comparison with other power systems. The high-pressure fluidized-bed boiler power system utilizes the potential advantages of fluidized-bed combustion over conventional combustion systems and has several potential advantages over an atmospheric fluidized-bed boiler system:

1. Boiler size is reduced. The potential for shop fabrication of utility boilers holds promise for large cost savings—both materials costs and field erection costs.
2. Power cycle efficiency can be increased.
3. Problems of solids handling and good distribution of fuel and air are reduced since the fluid bed area is small.
4. Scale-up problems are minimized.
5. Heat transfer surface is reduced by using a combined gas steam-turbine cycle.

Preliminary design calculations for a high-pressure fluidized-bed boiler power system are presented to illustrate the development of the concept and the identification of critical fluidized-bed combustor parameters. The design and evaluation of this concept are proceeding based on these parameters. Technical and economic evaluations may result in parameter changes in order to achieve an optimum design. Power plant efficiency and cost for the pressurized system have been calculated based on projected boiler efficiency and cost. The projected high-pressure system performance is compared with a conventional coal-fired power plant.

DESIGN BASIS

The development and evaluation of fluidized-bed boiler concepts must consider pollution control projections; market projections; power cost analyses; fluidized-bed combustor operating and design parameters; and power system operation, maintenance, and reliability.

Pollution Control Projections

Projected uncontrolled emissions from power plants and projected air-quality goals for SO_2, NO_x, and particulates have been considered.[1] Power plant design requirements for the year 2000, based on these projects, might be:

Emission	Control Level Leaving Stack
SO_2	100 ppm
NO_x	100 ppm
Particulates	0.010 gr/scf

Control targets for small particles (< 5 micrometers) have not been projected. However, regulations are anticipated which will require significant reductions from current emission levels. Information indicates that fluidized-bed combustors, operating with a coarse coal feed, may reduce small particle production by ∿ 60 wt percent.

Fluidized-bed boiler power systems have the potential for burning a broad range of low grade fuels, including solid wastes.

This provides a means for the effective use of natural resources and raw materials. Thermal discharge to cooling water may also be reduced if a pressurized combined gas steam cycle proves economic and operable

Market Projections

The impact of fluidized-bed combustion with sulfur absorption in the bed on air pollution abatement and on the economics of steam and power generation has been studied.[1] Results of the market studies have been used to establish fluidized-bed boiler specifications. These results are summarized elsewhere.[1]

Power Cost Analyses

Investment and operating costs projected for fluidized-bed boiler power systems must be compared between themselves and with conventional and proposed power systems. The fluidized-bed boiler power system must have an energy cost which is competitive with those of nuclear plants and fossil fuel plants with air pollution control. Present power plant investment costs are $186/kW for a 600-MW coal plant without pollution control and $269/kW for a 1100-MW nuclear plant.

Operating and Design Parameters

Operating and design variables for a fluidized-bed boiler system must be selected. Table 1 is a list of some of the important variables which must be considered. A given concept may not require consideration of all the parameters listed. However, careful consideration of these parameters, based on the analysis of available data and technology, is important if an optimum design is to be achieved.

Table 1. OPERATING AND DESIGN PARAMETERS FOR DESIGN OF FLUIDIZED-BED BOILERS

INDEPENDENT VARIABLES

Operating variables	Design variables
Fuel	Reactor configuration
Sulfur absorbent	Arrangement of boiler functions (e.g., economizer, superheater, regenerator, solids removal, carbon burnup cell)
NO_x control	Heat transfer surface (tube size, orientation, arrangement)
Bed temperature	Air distribution
Pressure	Solids handling
Gas velocity	Water steam flow
Excess air	Sulfur absorbent regenerator
Particle size	
Particle flow	
Bed height	

DEPENDENT VARIABLES

Operating effectiveness

Pollution control	Control
Combustion	Turndown
Sulfur sorbent utliization	System efficiency
Fluidization	Solids distribution
Heat transfer	Air distribution
Elutriation	Particle diffusivities
Attrition	Reliability
Corrosion	Pressure drop
Erosion	Heat release
Agglomeration	

Economics

- Tubing and fabrication
- Structures
- Controls
- Auxiliaries
- Water circulation
- Maintenance
- Steam headers
- Construction time
- Solids preparation and distribution
- Air preparation and distribution

Operation, Maintenance, and Reliability

Any new boiler design must consider the maintenance, operability, and reliability requirements of the system. The importance of these factors is clear in light of the present day brownouts and blackouts. The ability to achieve part-load operation with rapid response time is an important consideration for intermediate load generation which must vary in power production throughout the day.

A Basis for Evaluation

These constraints and guidelines provide a basis for the development, analysis, and evaluation of fluidized-bed boiler concepts. However, they must be viewed with flexibility. An example is the projected pollution control standards. Present data on the removal of sulfur dioxide in a fluidized-bed combustor desulfurizer is not adequate to permit the design of units to remove greater than approximately 90-95 percent of the sulfur. Thus, for a coal containing 4.3 percent S, it is not possible to design for control levels below ∿180 ppm with reliability until further data is available.

DESIGN CONCEPTS

Several boiler systems have been built, operated, or proposed which incorporate fluidized-bed combustion. Concepts go back to 1928 when Stratton developed a spouting fluidized-bed boiler.[2] The early boiler concepts incorporating fluidized-bed combustion were generally developed for burning low-grade fuels. These designs did not recover heat or consider sulfur removal in the fluidized-bed combustor. In the last decade, designs which incorporate heat recovery and sulfur removal in the fluidized-bed combustor have been conceived in England and in the United States.

In order to utilize the potential benefits of fluidized-bed combustion in minimizing air pollution while minimizing energy cost, new and existing concepts have been considered. Atmospheric and pressurized systems have been considered which incorporate various fluidized-bed configurations (single beds, stacked beds, unmixed beds, and segmented beds) and system component arrangements (solids feeding and removal, final carbon burnup, sorbent regeneration, heat transfer surface, and boiler functions).

One concept is the high-pressure fluidized-bed boiler combined gas steam cycle system which offers both large potential savings in investment and operating costs and effective pollution control. The concept illustrates the development and analysis of a fluidized-bed boiler system. Preliminary design calculations have been completed for the pressurized utility boiler system shown in Figure 1.

Power system performance studies and boiler cost and performance analyses for the system presented in Figure 1 show that the system should operate at 10 atm. using subcritical steam conditions. Preliminary design calculations have been made for a 600-MW pressurized utility boiler system operating at 10 atm with once-through subcritical steam. The operating conditions are summarized in Table 2.

Figure 2 is a process flow diagram for the pressurized utility boiler. The system consists of a primary fluidized bed combustor desulfurizer, cyclones for particulate removal, a carbon burnup cell, CBC, to achieve high combustion efficiency, and a limestone regenerator. The sulfur removal system being considered would feed regenerated stone from the regenerator to the primary fluidized bed. The regenerator is based on the system developed by Esso.[4] The high efficiency primary multicyclone removes 92.5 percent of the solids from the flue gas leaving the boiler. The collected solids are fed to the CBC, the combustion efficiency of which is 90 percent. The flue gas from the CBC is combined with the effluent from the primary cyclone and sent through the secondary multicyclone, which removes 97 percent of the solids, to the gas turbines. The combustion efficiency is approximately 98.5 percent; 1 percent of the carbon fed is assumed to be carried out in the flue gas and the CO concentration in the flue gas is ∿0.2 percent. Table 3 summarizes the material balance for the boiler and limestone regenerator. Figures 3 and 4 summarize the energy balance for the boiler and limestone regenerator.

Table 2. PRESSURIZED FLUID-BED BOILER POWER SYSTEM OPERATING CONDITIONS

Fluidized-bed combustor/desulfurizer			
Pressure	10 atm	Estimated limestone feed to achieve 95% removal	8 times stoichiometric CaO to react with SO_2
Temperature	1750°F		
Fuel	Pittsburgh No. 8 seam coal		
H.H.V	13,000 Btu/lb	Primary bed material	SO_2 removal agent
Excess air	10%	Solids elutriation	
SO_2 removal agent	Limestone	Ash	100%
		CaO/$CaSO_4$	∼0.5% of bed weight/hr
Carbon burnup cell			
Pressure	10 atm	Temperature	2000°F
Limestone regenerator			
Pressure	10 atm	Regeneration of $CaSO_4$	~90%
Temperature	2060°F		
Steam conditions			
Steam flow	3,500,000 lb/hr	Reheater inlet temp	650°F
Final feed-water temp	578°F	Reheater outlet temp	1000°F
Superheater outlet press.	2500 psi	Reheater inlet press.	600 psi
Superheater outlet temp	1000°F	Reheater outlet press.	580 psi
Reheat steam	3,200,000 lb/hr		
Gas turbine system			
Air temp leaving compressor	[illegible]36°F	Flue gas temp to lower stack gas cooler	525°F
Flue gas temp to gas turbine(s)	1600°F	Flue gas temp leaving system	275°F
Flue gas temp to upper stack gas cooler	831°F		

Table 3. MATERIAL BALANCE FOR PRESSURIZED UTILITY BOILER

Stream No.[a]	Description	Flow rate		Composition				Temperature °F
		Solids tons/hr	Gas moles/hr	Solids weight %		Gas mole %		
1-1	Ohio seam, Pittsburgh #8 coal	207	---	C	71.2	---		---
				H	5.4			
				O	9.3			
				N	1.3			
				S	4.3			
				Ash	3.5			
					100.0			
2-1	Air	---	160,000	---		N_2	77.5	636
						O_2	20.4	
						H_2O	2.1	
							100.0	
2-2	Air to FBCD	---	151,400	---		Same as 2-1		636
2-2A	Air to CBC	---	8,600	---		Same as 2-1		636
2-3	Flue gas from FBCD	29.4	158,900	Ash	59.9			~1750
				C	29.9			
				Spent limestone	10.2			
					100.0			
2-3A	Flue gas from CBC	19.9	8,600	Ash	81.9			~2000
				C	4.0			
				Spent limestone	14.1			
					100.0			
2-4	Solids to CBC from primary cyclone(s)	27.2	---	Same as 203		---		~1700

Table 3 (continued). MATERIAL BALANCE FOR PRESSURIZED UTILITY BOILER

Stream No.[a]	Description	Flow rate		Composition				Temperature °F
		Solids tons/hr	Gas moles/hr	Solids weight %		Gas mole %		
2-5	Flue gas from primary cyclone(s)	2.2	158,900	Same as 2-3				~1700
2-6	Combined flue gas to secondary cyclone(s)	22.1	167,500	Ash C Spent limestone	79.7 6.6 13.7 100.0	N_2 CO_2[b] CO H_2O O_2 NO SO_2	74.2 15.0 0.2 8.7 1.8 ~400 ppm ~170 ppm 100.0	~1600
2-7	Flue gas to gas turbine(s)	0.66 (0.15 gr/SCF)	167,500	Same as 2-6		Same as 2-6		1600
2-8	Waste solids	21.4	---	Same as 2-6		Same as 2-6		1600
3-1	Regenerated limestone to FBCD	139.3	---	CaO $CaCO_3$ $CaSO_4$	77.4 20.4 2.2 100.0	---		
3-2	Spent limestone from FBCD	145.6	---	CaO $CaSO_4$	73.5 26.5 100.0	---		~1750
3-3	Regenerated stone from regenerator	123.3	---	CaO $CaSO_4$	97.2 2.8 100.0	---		~2000
3-4	Waste stone	12.3	---	Same as 3-3		---		

Table 3 (continued). MATERIAL BALANCE FOR PRESSURIZED UTILITY BOILER

Stream No.[a]	Description	Flow rate		Composition				Temperature °F
		Solids tons/hr	Gas moles/hr	Solids weight %		Gas mole %		
3-5	Makeup limestone	28.4	---	$CaCO_3$	97.0	---		
4-1	Coke	9.2	---	C Ash	98.0 2.0 100.0	---		
5-1	Air	---	6,070	---		Same as 2-1		
5-2	SO_2 rich flue gas	2.2	6,800	CaO $CaSO_4$ Ash	72.7 18.2 9.1 100.0	N_2 CO_2 CO SO_2	70.1 22.4 negligible 7.5 100.0	~2000

[a] See Figure 2.

[b] Includes CO_2 produced from calcining makeup limestone.

Figure 5, a temperature-enthalpy diagram for the boiler, graphically represents bed temperatures, water and steam temperatures, estimated boiler tube-wall temperatures, and the energy breakdown of the system. Table 4 summarizes the heat transfer surface requirements for the boiler and estimates of the tube-wall temperatures. The maximum tube wall temperature estimate is 1150°F. These calculations are based on a bed-to-tube heat transfer coefficient of 50 Btu/hr-ft^2-°F, which is considered pessimistic.

Table 4. PRESSURIZED FLUID-BED BOILER OPERATING CONDITIONS AND DESIGN PARAMETERS

Function	Pre-evaporator		Evaporator	Superheater	Reheater	Total
	FBCD	CBC				
Heat transferred, Btu/hr x 10^8	6.3	1.2	8.8	14.1	6.1	36.5
Overall heat transfer coeff, Btu/hr-ft^2-°F	47	47	47	43	40	--
Surface requirements, ft^2	12,000	1,900	17,200	36,400	16,700	84,200
Inlet tube wall temperature estimates, °F	660	750	730	820	870	–
Outlet tube wall temperature estimates,° F	730	750	730	1110	1150	–

Table 5 summarizes the performance of the pressurized-bed boiler power system. The net power production is 635 MW, with the steam cycle producing ∿85 percent of the power. The boiler efficiency, 88.6 percent, is based on conventional procedures for calculating boiler performance. A term has been added to account for the loss from sensible heat of the spent limestone leaving the bed. Pollution control requirements for the preliminary design concentrated on sulfur removal. A basis of 95 percent removal was selected for the preliminary design since reliable data is not available to design for sulfur removal greater than 90-95 percent. Techniques for reducing nitrogen oxides have not been considered in the initial design. Design specification for the particulate removal system after the fluidized-bed boiler is based on particulate loading requirements for the gas turbine. Results from the BCURA high-pressure boiler erosion tests have been used as the basis.[4] Their results indicate that particulate loadings of 0.15 gr/scf may be permitted in the gas to the turbine. Additional tests and analyses are required to determine the effect of coal ash and spent limestone or other SO_2 sorbent on turbine blade life. Final dust removal would be added after the gas is cooled to achieve the 0.01 gr/scf stack emission level.

Details of the fluidized-bed combustor desulfurizer design have been considered based on the operating conditions and design parameters. A shop-fabricated modular design is being considered for the high-pressure boiler in order to achieve reduced boiler erection time and added savings in construction costs. Rail shipment requirements are approximately 12 x 16 x 40 ft. With a vertical unit, the active fluidized-bed cross-sectional area can be 50-60 ft^2. The number of modules and the bed depths will be determined by the heat transfer surface requirements, heat transfer surface configurations, operating and structural considerations, and cost. Table 6 shows that the effect of tube bundle design on the volume requirement is important for large utility boilers. If a single vertical deep-bed concept were adopted for each module and if the tube bundle design consisted of horizontal 1-in. tubes on a 2-in. triangular pitch, the boiler would require five shop-fabricated vessels with 20-30 ft deep beds, each serving a separate function. If a stacked bed design were adopted with each module containing each of the respective boiler functions, six vessels would be required; the extra vessel would be required because of the increased space requirements for gas distributors and freeboard. The number of vessels required for each concept is doubled by going to 2-in. tubes with a 4-in. pitch. The boiler would then require 10-12 vessels. With this design, shop fabrication must be weighed with field erection of a larger unit(s). Construction costs, external piping, solids handling, and control problems must be studied in order to determine the best system. The optimum heat transfer arrangement will be selected based on steam-side pressure drop, tube costs, header design, and fluidization, in addition to the effect of boiler tube configuration on bed volume.

Table 5. PRESSURIZED FLUID-BED BOILER POWER SYSTEM PERFORMANCE

Power	
Steam turbine	538.4 MW
Gas turbine	113.1 MW
Plant requirement	16.1 MW
Net power	635.4 MW
Efficiency	
Plant heat rate	8975 Btu/kWhr
Plant efficiency	38%
Boiler system losses[a]	
Dry gas (based on stack temperature)	3.88%
Hydrogen and moisture in fuel	4.14%
Moisture in air	0.08%
Unburned combustible	1.51%
Radiation	0.15%
Sensible heat of solids	0.11%
Unaccounted for losses	1.50%
Total losses	11.37%
Boiler efficiency	88.6%
Pollution control	
SO_2 in stack gas	160 ppm[b]
Particulate loading to gas turbine(s)	0.15 gr/scf

[a] Based on heats of reaction for coal-O_2 and CaO-SO_2; ∿13300 Btu/lb coal.

[b] Emission is less than that from boiler due to air added to gas turbine system for cooling.

Variation in pressure drop through the boiler system has little effect on plant performance for the pressurized power system. A 1-percent increase in the pressure drop through the boiler system results in a 0.16-percent decrease in power and a 0.09-percent increase in heat rate. This permits the use of deep beds, and high pressure drop distribution plates and dust collection equipment without significant loss in efficiency.

Under contract to Westinghouse, Foster Wheeler is providing (for NAPCA) proposal drawings of both a pressurized and an atmospheric utility boiler. Foster Wheeler will describe their work in a companion paper.

DESIGN COMPARISON

Fluidized-bed boiler designs and power systems must be compared between themselves and with conventional boiler and power systems. The pressurized boiler power system concept is used to illustrate the types of comparisons which must be considered.

Comparison of Fluidized-Bed Boiler Designs

Fluidized-bed boiler concepts are compared on the basis of pollution control effectiveness, cost, efficiency, and operating characteristics. The variable space which must be considered in developing a fluidized-bed boiler design was discussed and summarized in Table 1. To achieve an optimum design, the alternatives available for each variable will require analysis and comparison.

Selection of a fluidized-bed configuration for the pressurized boiler is an example of a critical design task which is being evaluated. Several configurations offer promise: vertical units with single deep beds, vertical units with stacked beds (each fulfilling a separate boiler function), open horizontal beds, and segmented horizontal beds. Each concept can be considered for shop fabrication or field erection. A comparison of these configurations must be made based on operating characteristics (turndown, solids handling, air distribution, erosion, etc), heat transfer surface design, sulfur removal and sorbent regenerator design, freeboard requirements, fluidization, combustion efficiency, and cost. For example, single deep (on the order of 30 ft) beds offer several potential advantages: solids handling problems can be reduced (feeding and removal), the number of air distributor plates is reduced, available space is used effectively, heat transfer surface out of the bed is minimized, higher combustion efficiency may be achieved as the result of longer residence time*, and boiler size and cost can be reduced. Problem areas can also be visualized: coal distribution through the bed, temperature profile through the bed, vibration, startup and turndown capability, and tube maintenance. Stacked vertical beds offer a method of providing the four boiler functions in each module which may simplify startup and turndown of the system. The horizontal configuration simplifies structural

Table 6. FLUIDIZED-BED VOLUME REQUIREMENT FOR HEAT TRANSFER SURFACE

Tube arrangement	Triangular configuration			Rotated-square configuration
	1-in. diam, 2-in. pitch	1-in. diam, 3-in. pitch	2-in. diam, 4-in. pitch	2-in. diam, 4-in. pitch
Pre-evaporator volume, ft^3	1,280	2,880	2,560	2,950
Evaporator volume, ft^3	1,580	3,560	3,160	3,650
Superheater volume, ft^3	3,340	7,530	6,700	7,720
Reheater volume, ft^3	1,530	3,450	3,070	3,540
Total volume, ft^3	7,730	17,420	15,490	17,860

---

* Deep beds may be able to achieve the high combustion efficiencies with coarse particles that have been projected for fine particles in shallow beds.

design problems and offers alternate methods for achieving turndown and recycling solids. This configuration does not use the volume as efficiently as does the vertical unit and may also limit the flexibility in heat transfer surface design. Fuel distribution may be more difficult over the larger area in a horizontal design. If lower gas velocities are used, carbon carryover may be reduced; although deeper beds with vertical units may provide the same result. These comments illustrate the types of comparative analyses which must be considered before selecting a configuration.

The achievement of high combustion efficiency is another major concern. A vertical deep-bed design may permit solids to be recycled back to the bed as a means for achieving high combustion efficiencies. With shallow beds and gas velocities greater than 2-4 fps, the CBC concept conceived by Pope, Evans and Robbins[5] has marked advantages. With a deep bed, the possibility exists for achieving high efficiencies at high velocities by recycling material. The tradeoffs which must be considered are solids handling problems, operating cost, and control. It does not appear practical to separate the CBC from the pressurized boiler unit since this would involve additional transport of solids in and out of high-pressure vessels. Several CBC concepts have been conceived to accomplish this goal. Since the CBC represents a small fraction of the total bed volume (see Table 4), it is not practical to allow a single bed in each module for the CBC. A single CBC could be designed for one module and solids transferred from the other modules; however, this increases the solids handling problem and is not recommended. Although a single high-pressure field-erected unit might reduce the problems with a single CBC, it is not recommended as a solution. Thus, design configurations have been considered which incorporate the CBC in a section of one of the beds in a stacked bed design; e.g., in a quadrant of the bottom bed. Solids are then transferred to the CBC from the upper beds through an internal cyclone(s).

The design of the sulfur removal system illustrates another area where critical analysis is required. Processes for high-temperature/high-pressure regeneration of the sulfur removal agent must be considered. Given a regeneration process, the location must be considered. If the regenerator is external to the fluidized bed, solids must be transported at high temperature and pressure to the regenerator and the regenerated solids must be returned to the boiler. Incorporating the regenerator in the high-pressure fluidized-bed boiler may greatly reduce the solids handling problem, minimize heat losses, and reduce space requirements. An additional process scheme which may offer advantages is to combine the carbon burnup cell (CBC) and the regenerator into a single unit. Both systems operate near 2000°F and the energy requirements for the regenerator can be supplied by the solids to the CBC. (See Figures 3 and 4.) If the CBC is operated with 30 percent excess air, the excess air may reduce the SO_2 concentration in the flue gas from the regenerator-CBC to a level which cannot be handled by a sulfur recovery plant. The carbon monoxide requirements for efficient operation of the regenerator must be studied as well as the potential problems of matching the operation of the CBC, regenerator, and the fluidized-bed combustor/desulfurizer.

Solids feeding (tangential, bottom, side, and top), solids removal, and arrangements of boiler functions and heat transfer surface configurations (vertical, horizontal, spiral, and serpentine) are being studied and evaluated. A range of each operating condition must be considered and the tradeoffs evaluated: e.g., the effect of bed temperature on corrosion, deposits, SO_2 absorption, NO_x emissions, combustion efficiency, heat transfer surface requirements, and ash fusion; and the effect of particle size on kinetics, combustion efficiency, heat transfer, elutriation, and preparation costs. Higher plant efficiencies may also be possible since steam temperature may not be limited by corrosion and deposits.

These examples are presented to illustrate the types of analyses and comparisons which must be made between fluidized-bed boiler designs. They are by no means complete.

Comparison with Conventional Systems

The high-pressure boiler requires the analysis of a total power system. Thus, comparison with conventional systems must ultimately be based on overall power plant performance. The high-pressure FBB power system is compared with a conventional boiler and power system on the basis of common heat transfer surface, efficiency, cost, operating characteristics, and pollution control.

Heat Transfer Surface. The estimated heat transfer surface for the pressurized boiler is compared with the surface requirements for the Hammond Unit No. 4 which has essentially identical steam conditions. The surface requirements for the pressurized boiler are approximately 30 percent of those for a conventional coal-fired boiler. Table 7 presents the comparison.

Efficiency. The projected pressurized FBB efficiency of 88.6 percent compares favorably with the 89 percent of the Hammond Unit No. 4. The pressurized FBB power efficiency is 38 percent, 1 percent greater than that of the Hammond unit. Higher efficiencies could be achieved with the pressurized boiler system by increasing the gas temperature to the gas turbine. This could be achieved by modifying the system to produce some fuel gas which can be combusted at high temperature and mixed with the low-temperature fluidized-bed combustor gas. If the combustor were operated at a higher temperature, the sulfur removal process would not be effective.

Cost. The projected plant cost (on an installed basis for plant operation in 1975) for the pressurized power plant is $158/kW. This is based on a boiler efficiency of 89 percent, a fluidized-bed boiler/desulfurizer system cost of $40/kW of steam turbine power, and a 15-

Table 7. COMPARISON OF HEAT TRANSFER SURFACE AREA

Function	Heat transferred, Btu/hr x 10^8		Heat transfer Surface, ft^2			Surface reduction with pressurized boiler %
	FW Hammond unit No. 4 (3,600,000 lb steam/hr)	Pressurized fluid-bed boiler (3,500,000 lb steam/hr)	FW Hammond unit No. 4		Pressurized fluid-bed boiler	
			Reported	Total surface estimated[a]		
Pre-evaporator	7.8[b]	7.5	25,900	81,000	13,900	62
Evaporator	9.1	8.8			17,200	
Superheater	14.5	14.1	81,600[c]	118,000	36,400	69
Reheater	6.3	6.1	113,000	109,000	16,700	85
	——	——	——	——	——	—
	37.7	36.5	220,500	308,000	84,200	73

[a] Converts surface reported as projected surface to actual tube surface.

[b] Based on temperature leaving economizer of 593° F.

[c] Surface includes both projected and actual.

percent reduction in indirect costs. A boiler system cost of $40/kW (steam) does not appear unreasonable for a modularized design, which also permits indirect cost savings on construction and interest during construction. A conventional 600-MW coal-fired power plant cost in 1970 dollars is estimated to be $186/kW, without pollution control equipment for sulfur removal.

Turndown. The modular design of a pressurized boiler power system is projected to have the capability of achieving turndown ratios of 4:1 or greater. This compares favorably with turndown ratios of conventional coal-fired plants.

Pollution Control. The pressurized-boiler power system offers advantages for controlling sulfur and particulate emissions. Although control of nitrogen oxide emissions has not been considered in the present design, techniques are being studied to lower them. The pressurized-boiler was designed for 95 percent sulfur removal. Data is limited which permits design calculations to be made at the 95-percent level. Thus, refinement of the designs to achieve high SO_2 removal may be required. Particulate removal from the pressurized system has not been considered beyond the gas turbines. Particle size distributions of emissions from fluidized-bed combustors indicate that the emission of particles <5 micrometers may be significantly less than those from conventional units. Conventional coal-fired power plants using a similar coal have SO_2, particulate, and NO_x emissions of approximately 3200 ppm, 0.2 gr/SCF, and 500 ppm, respectively.

Table 8 summarizes a comparison of the preliminary design of a pressurized-boiler power system with a conventional coal-fired power system. Based on the preliminary design and analysis of a pressurized fluid-bed boiler power plant, such a system has the potential for providing not only compact, cheap, and efficient power, but also clean air and effective use of our natural resources.

CONCLUSIONS

Fluidized-bed boiler concepts are being generated and evaluated for cheap and efficient power generation with air pollution control. A broad range of fluidized-bed combustor operating and design parameters are being studied in order to achieve an optimum design. The concepts are developed and compared with other fluidized-bed boiler systems and with conventional power systems, based on: projections for emission regulations of SO_2, NO_x, and particulates; the power generation market; boiler and power plant costs; and boiler and power system operation, maintenance, and reliability.

A high-pressure fluidized-bed boiler power system illustrates the development and evaluation of a concept. Power system operating conditions have been specified, preliminary engineering calculations have been performed, critical fluidized-bed combustor parameters have been identified, and proposal designs and evaluations are underway. Preliminary comparisons have been made with conventional power systems. Data indicates that sulfur emissions can be reduced 95 percent. The fluidized-bed boiler also offers advantages in controlling particulates and may reduce NO_x emissions. The amount of heat transfer surface required by a pressurized boiler operating in conjunction with a combined gas/steam turbine power cycle is estimated to be ∿30 percent of that required by a conventional boiler for the same steam conditions and flow. The preliminary cost estimate for the fluidized-bed boiler power system is $158/kW. The cost estimate for a conventional coal-fired power plant, allowing $10/kW for SO_2 removal, is $196/kW. The projected efficiency of the fluidized-bed boiler is 88.6 percent, which compares favorably with a conventional plant of the same capacity. Thus, the high-pressure fluidized-bed boiler power systems shows potential technical and economic advantages over a conventional coal-fired power plant.

Table 8. COMPARISON OF PRESSURIZED-BOILER POWER PLANT WITH CONVENTIONAL POWER PLANT

Function	Conventional coal-fired plant	Pressurized fluid-bed boiler power plant
Boiler tube surface, ft^2	308,000	84,200
Efficiency		
Boiler, %	89.0	88.6
Plant, %	37.0	38.0
Cost, $/kW	186[a]	158
Turndown	4:1	4:1
Pollution control		
SO_2, ppm	∿3200	160
Particulate		
Total, gr/SCF	∿0.2	<0.15[b]
5 µm, %	15	∿5
NO_x, ppm	>500	<250

[a] 600-MW plant without pollution control equipment.

[b] Particulate removal beyond the gas turbine requirements has not been considered in the initial design.

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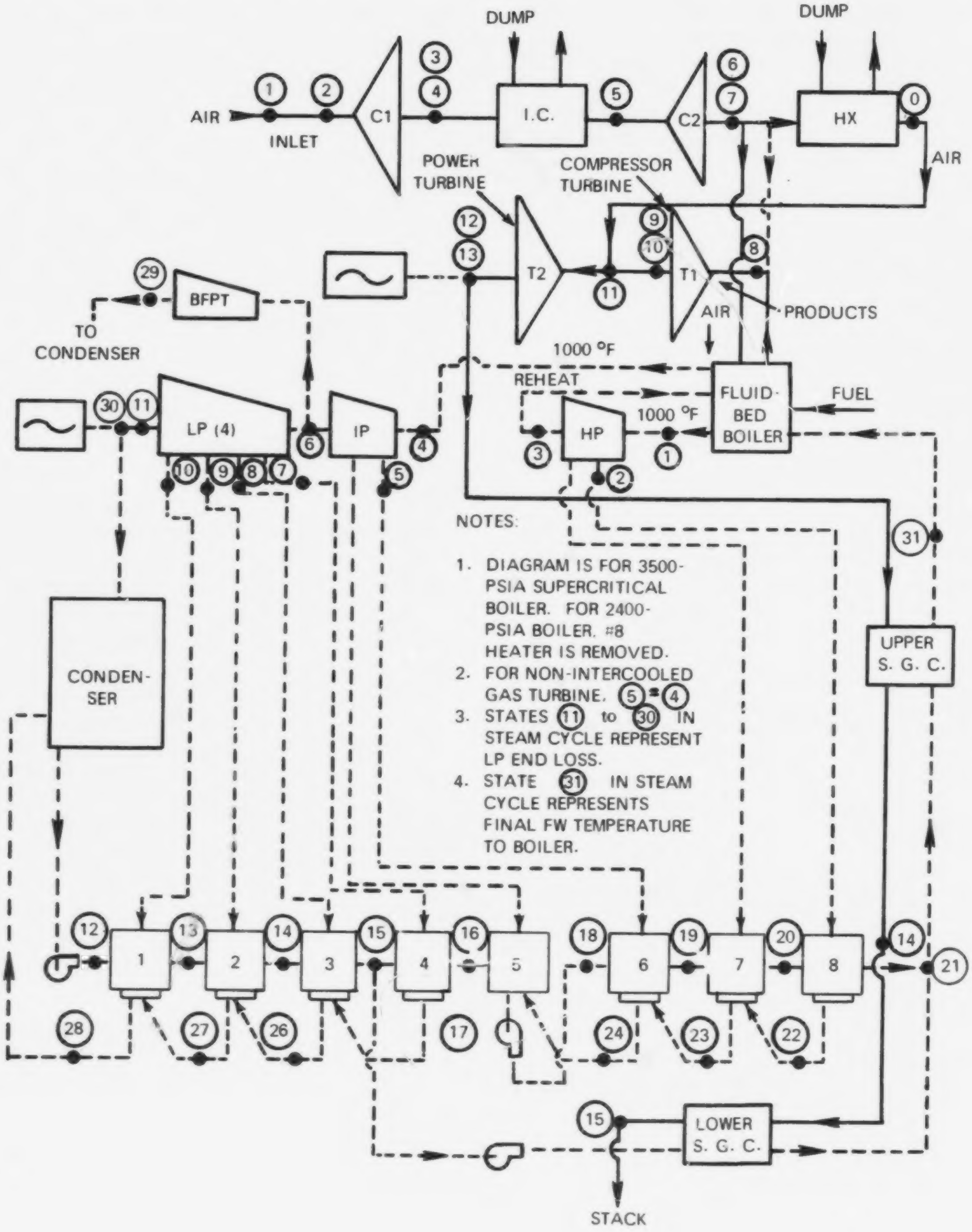
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AIR
INLET
①
②
C1
③
④
I.C.
⑤
C2
⑥
⑦
HX
⓪
AIR
POWER TURBINE
COMPRESSOR TURBINE
⑫
⑬
T2
⑨
⑩
T1
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⑧
AIR
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BFPT
TO CONDENSER
1000 °F
REHEAT
FLUID-BED BOILER
FUEL
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LOWER S. G. C.
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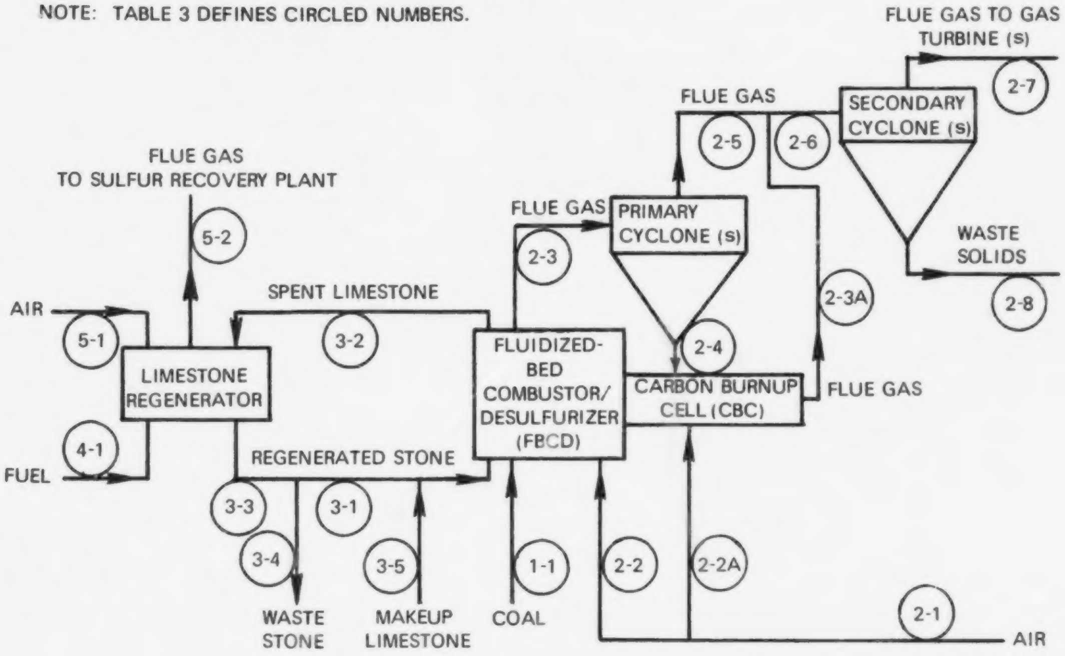
NOTES:

1. DIAGRAM IS FOR 3500-PSIA SUPERCRITICAL BOILER. FOR 2400-PSIA BOILER, #8 HEATER IS REMOVED.
2. FOR NON-INTERCOOLED GAS TURBINE, ⑤ = ④
3. STATES ⑪ to ㉚ IN STEAM CYCLE REPRESENT LP END LOSS.
4. STATE ㉛ IN STEAM CYCLE REPRESENTS FINAL FW TEMPERATURE TO BOILER.

Figure 1. High-pressure fluid-bed boiler power system (block diagram, combined cycle).

NOTE: TABLE 3 DEFINES CIRCLED NUMBERS.



FLUE GAS TO GAS TURBINE (s)
2-7
FLUE GAS
2-5
2-6
SECONDARY CYCLONE (s)
FLUE GAS TO SULFUR RECOVERY PLANT
5-2
FLUE GAS
PRIMARY CYCLONE (s)
2-3
WASTE SOLIDS
2-3A
2-8
AIR
SPENT LIMESTONE
5-1
3-2
FLUIDIZED-BED COMBUSTOR/ DESULFURIZER (FBCD)
2-4
LIMESTONE REGENERATOR
CARBON BURNUP CELL (CBC)
FLUE GAS
4-1
FUEL
REGENERATED STONE
3-3
3-1
3-4
3-5
1-1
2-2
2-2A
2-1
WASTE STONE
MAKEUP LIMESTONE
COAL
AIR

Figure 2. Process flow diagram for pressurized utility boiler.

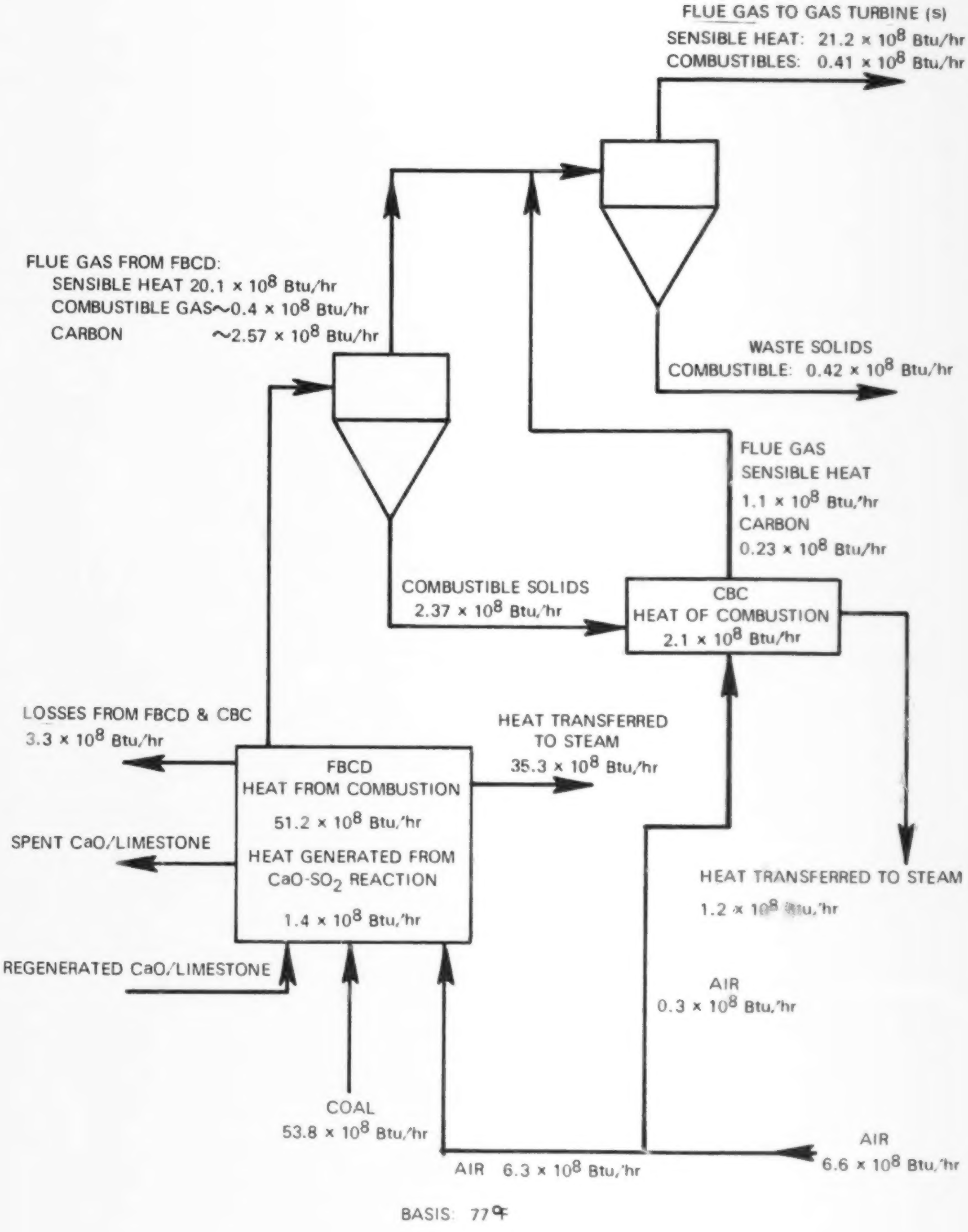

FLUE GAS TO GAS TURBINE (s)
SENSIBLE HEAT: 21.2 × 10⁸ Btu/hr
COMBUSTIBLES: 0.41 × 10⁸ Btu/hr
FLUE GAS FROM FBCD:
SENSIBLE HEAT 20.1 × 10⁸ Btu/hr
COMBUSTIBLE GAS ~0.4 × 10⁸ Btu/hr
CARBON ~2.57 × 10⁸ Btu/hr
WASTE SOLIDS
COMBUSTIBLE: 0.42 × 10⁸ Btu/hr
FLUE GAS
SENSIBLE HEAT
1.1 × 10⁸ Btu/hr
CARBON
0.23 × 10⁸ Btu/hr
COMBUSTIBLE SOLIDS
2.37 × 10⁸ Btu/hr
CBC
HEAT OF COMBUSTION
2.1 × 10⁸ Btu/hr
LOSSES FROM FBCD & CBC
3.3 × 10⁸ Btu/hr
HEAT TRANSFERRED
TO STEAM
35.3 × 10⁸ Btu/hr
FBCD
HEAT FROM COMBUSTION
51.2 × 10⁸ Btu/hr
HEAT GENERATED FROM
CaO-SO₂ REACTION
1.4 × 10⁸ Btu/hr
SPENT CaO/LIMESTONE
REGENERATED CaO/LIMESTONE
HEAT TRANSFERRED TO STEAM
1.2 × 10⁸ Btu/hr
AIR
0.3 × 10⁸ Btu/hr
COAL
53.8 × 10⁸ Btu/hr
AIR 6.3 × 10⁸ Btu/hr
AIR
6.6 × 10⁸ Btu/hr
BASIS: 77°F

Figure 3. Energy balance for pressurized utility boiler.

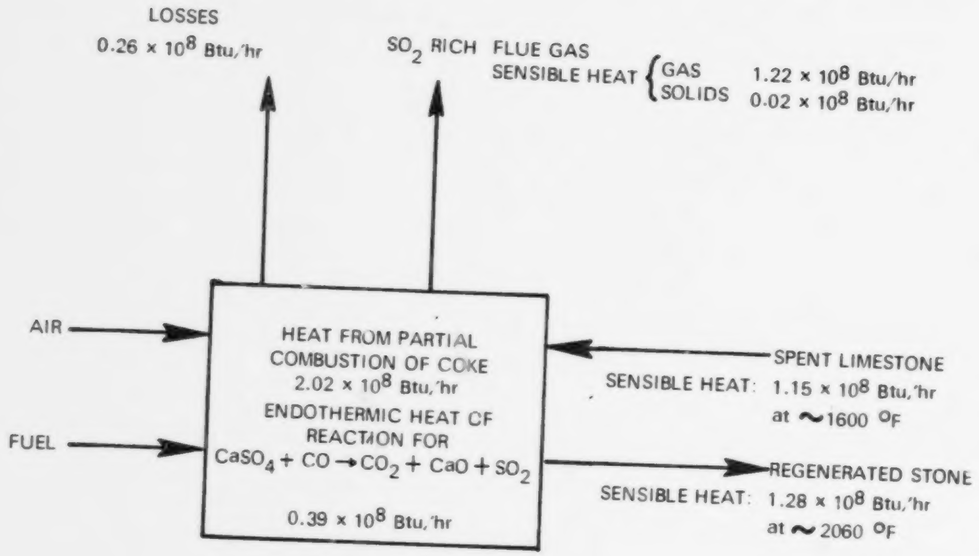

LOSSES
0.26 × 10^8 Btu/hr
SO2 RICH FLUE GAS
SENSIBLE HEAT
GAS 1.22 × 10^8 Btu/hr
SOLIDS 0.02 × 10^8 Btu/hr
AIR
FUEL
HEAT FROM PARTIAL COMBUSTION OF COKE
2.02 × 10^8 Btu/hr
ENDOTHERMIC HEAT OF REACTION FOR
CaSO4 + CO → CO2 + CaO + SO2
0.39 × 10^8 Btu/hr
SPENT LIMESTONE
SENSIBLE HEAT: 1.15 × 10^8 Btu/hr
at ~1600 °F
REGENERATED STONE
SENSIBLE HEAT: 1.28 × 10^8 Btu/hr
at ~2060 °F

Figure 4. Energy balance for limestone regenerator.

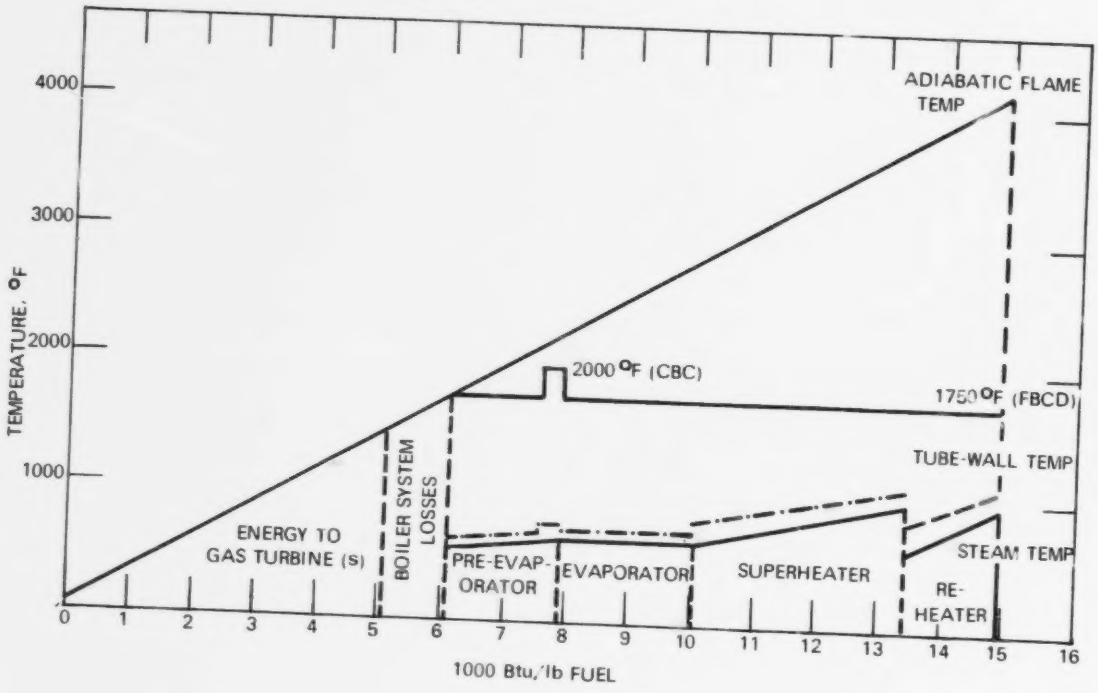


Figure 5. Temperature-enthalpy diagram for pressurized utility boiler.

SESSION V:

Conceptual Design and Economic Feasibility (Continued)

SESSION CHAIRMAN:

Mr. T. C. L. Nicole. National Coal Board, England

1. DEVELOPMENT OF FLUIDISED-BED COMBUSTION FOR FIRING UTILITY STEAM BOILERS

D. H. BROADBENT

National Coal Board, England

The national Coal Board's interest in fluidised-bed combustion stems from a desire to maintain its coal sales to its largest outlet. electricity generation, against growing competition from other sources of energy.

Figure 1 is a projection of the United Kingdom energy market up to the end of the century. It reflects the most optimistic forecasts available for the part to be played by nuclear energy and natural gas, yet shows an increasing use of fossil fuels from 1975 on. Hence the Board's justification for research and development of fluidised-bed combustion as a possible means of reducing the capital and running costs of fossil-fuel-fired boilers.

We have compared our energy forecasts with those for the U.S.[1] which show a similar trend up to 2000 A.D.; but in the U.S., there is the added incentive of pollution control which makes fluidised-bed combustion even more attractive.

Work at both National Coal Board laboratories (B.C.U.R.A. and C.R.E.) has proceeded for 7 years and costs are currently running at £1/2 ($1-1/4) million per annum. We have 94 scientists working on the project, backed by an engineering workshop and other supporting staff.

There are 11 major research tools: 7 hot and 4 cold: 10 at atmospheric pressure and 1 pressurised to 6 atmospheres. Results from these rigs have been used to construct a mathematical model from which the parameters of a variety of boiler designs have been established.

In addition, some £200,000 has been spent over the last 2 years in obtaining designs of 20-, 120-, and 660-MW boilers from manufacturers of international standing. In all cases, at least two designs have been obtained for each boiler size from different contractors to ensure unbiased designs. N.C.B. owns these contractors' designs exclusively.

The programme of development shown in Figure 2 covers both pressurised and atmospheric units and is planned at a rate which will enable building of a 660-MW unit in as short a time as possible without risking excessive scaleup factors. The programme covers a 19-year period, 7 of which have gone.

The next major capital investment is on the Grimethorpe boiler, planned for one of the Board's power stations in Yorkshire. It is 20 MW with a 12- x 36-ft bed and will take 12-18 months to build and commission. The boiler, designed in detail, was sent out to tender to six firms: five offers were received, each gave a design to specification and also their own version. Results of bids were remarkably close, and the Board is now in position to place a firm contract. Eighteen months of experimentation is planned for the Grimethorpe boiler, by which time we would have a 120-MW design out for tenders. A 120-MW boiler takes some 3 years to build and commission and would then require 2 years of testing. During this 2-year period, a 660-MW design would be put out for tenders, and would take 5 years for construction and commissioning.

Total cost of this programme is some £45 million (at 1969 prices) over the 19-year period. Our market surveys indicate that there would be considerable offsetting of these costs before the end of the 19-year period by virtue of specific experimental work we will carry out for other people, commercial exploitation of boilers in the 20-MW range possible from 1973, and similar exploitation of boilers in the 120-MW range from 1978.

If pressurised fluidised-bed combustion is proceeded with alone, it would follow a similar pattern; but the time scale would also be influenced by time requirements for suitable industrial gas turbine developments.

We would aim to run the pressurised and atmospheric programmes concurrently in which case the information crossflow will ensure some considerable cost sharing.

The question now is when does the N.C.B. stop going it alone in the U.K. and start to attract financial support from others. We have decided to seek partners for the Grimethorpe stage.

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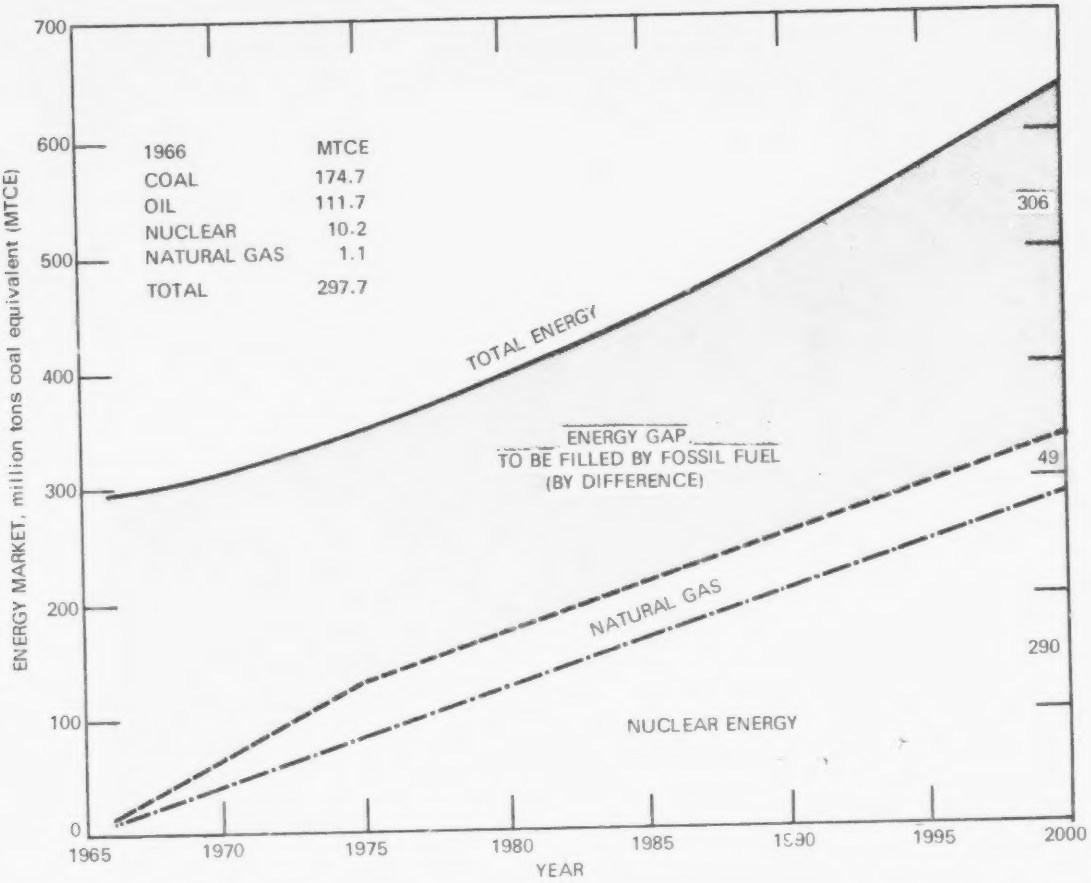

Figure 1. Projections showing the total fossil fuel "energy gap" up to the year 2000.

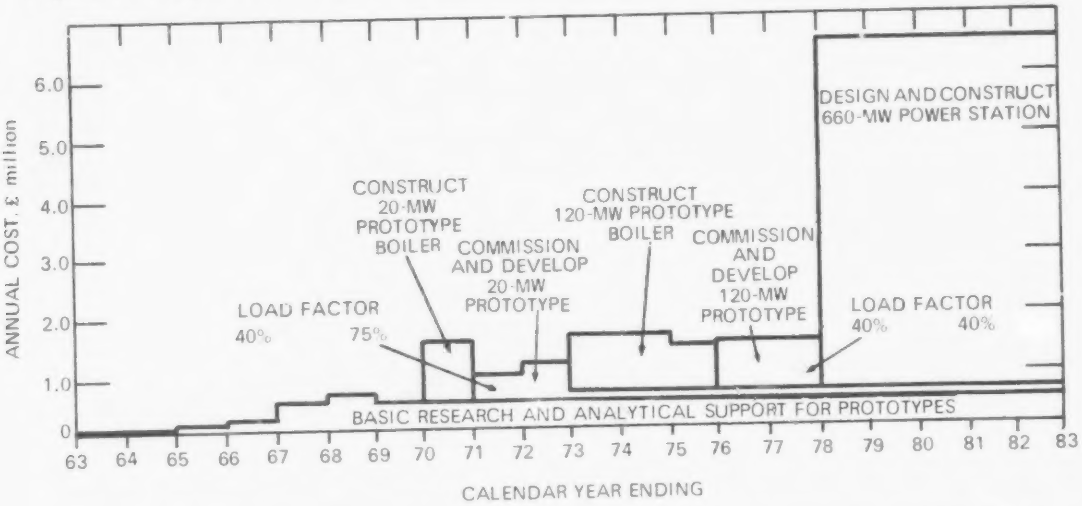

Figure 2. Cost and time scale of developing fluidised-bed combustion for central power station boilers (present day prices).

2. MARKETABLE DESIGNS FOR FLUIDIZED - COMBUSTION BOILERS

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INTRODUCTION

Westinghouse, under contract to NAPCA, is providing market studies, hardware designs, performance evaluations, and economic data for fluidized-bed boilers suited to industrial and to utility applications. Recommendations are also being furnished regarding the development program and pilot plant operation required to realize practical boiler systems which are effective in air pollution control and economic in operation.

MARKET STUDIES

The industrial and utility boiler market surveys, which are nearing completion, have two purposes:

1. To determine the possible impact of fluidized-bed combustion with sulfur absorption in the bed both on air pollution abatement and on the economics of steam and power generation.
2. To establish functional specifications for the fluidized-bed boiler designs that are responsive to customer needs.

Industrial Boiler Installations

Table 1 shows historic data for the industrial water-tube boiler market; statistics are being projected so that boiler sales can be predicted through 1980. Certain trends are already clear:

1. The total capacity of industrial boilers installed each year is increasing.
2. The number of new coal-fired installations are sharply decreasing and gas-fired installations are increasing.
3. The average capacity of new boilers has increased from about 60,000 lb/hr to 75,000 lb/hr in the past 7 years. (Coal-fired boilers are 30-60 percent larger than the average.)

The operating life of an industrial boiler is about 30 years. A conservatively low estimate of the total industrial boiler capacity now extant is 1.2×10^9 lb/hr; total annual fuel costs for these might be about $2.5 billion.

The possible pollution control and economic benefits of developing a successful fluidized-bed combustion industrial boiler are difficult to assess. Purchasers of industrial boilers currently are avoiding SO_2 and particulate (but not NO_x) emission problems by using natural gas or low sulfur oil. But the supplies of such fuels are limited and their costs can be expected to rise. (These points will be expanded later.) In certain areas of the U.S. it has been announced that available natural gas is inadequate for new industrial customers. There may well be a growing need for low-cost packaged industrial boiler in sizes up to 250,000 lb/hr (perhaps even larger) that will burn coal in such a way as to minimize SO_2, NO_x, and particulate emissions. Our preliminary design work indicates that the fluidized-bed boiler is indeed more compact than the conventional coal-burning boiler and

Table 1. ANNUAL SALES OF INDUSTRIAL WATER-TUBE BOILERS IN THE U.S.

	1963	1965	1968	1969
Total capacity sold, lb/hr x 10^{-6}	50.5	76.9	67.0	78.0
Coal-fired	10.0	12.1	4.1	2.1
Oil-fired	12.3	17.4	15.6	13.3
Gas-fired	20.2	34.6	39.6	47.8
Other[a]	8.0	22.8	7.7	14.8
Total number sold	879	1055	908	
Coal	125	104	32	
Oil	263	296	194	
Gas	409	561	617	
Other[a]	82	94	65	

FOB costs of a packaged industrial boiler

Coal	(up to 60,000 lb/hr)	\$1.25 per lb/hr
	(above 60,000 lb/hr field erection)	\$3 to \$4 per lb/hr
Oil	(up to 250,000 lb/hr)	\$0.80 to \$1.00 per lb/hr
Gas	(up to 250,000 lb/hr)	\$0.80 to \$1.00 per lb/hr

[a] Includes bagasse, black liquor, bark, and waste.

thus can be packaged in sizes up to 250,000 lb/hr. Emissions of SO_2 can be reduced by a solid absorbent–such as CaO or dolomite–in the bed, or it may well be cheaper to use a desulfurized char fuel. Use of such a fuel would eliminate (or minimize) the use of a solid absorbent and the problem of disposing of it or regenerating it. Fluidized-bed combustion may also be effective in reducing NO_x because of its lower operating temperature. Particulates may also be more readily removed from stack gases; they are, in general, larger than particles from pulverized fuel combustion; because they are not sintered, wear of cyclones and mechanical collectors should be minimized; and their high carbon content may improve the operation of electrostatic precipitators.

Fluidized-bed industrial boilers may well be effective and economic in air pollution control if natural gas and low sulfur oil rise in cost and if supplies of a desulfurized char become readily available. The costs of operating an industrial boiler with a sulfur absorbent in the bed, regenerating the absorbent, and recovering (or disposing of) the sulfur will be assessed to determine the economic feasibility of using a high sulfur coal (or oil).

Utility Boiler Installations

Table 2 predicts the total generating capacity and new installations of generating capacity in the U.S. The new installations are broken down into:

1. Base load plants–with a load factor greater than 80 percent and employing fossil fuel and a boiler.
2. Intermediate load plants–with a load factor around 45 percent and employing fossil fuel and a boiler.

Table 2. ANNUAL SALES OF UTILITY STEAM GENERATORS IN THE U. S. AND ELECTRIC UTILITY POWER GENERATION

	1970	1975	1985
Total installed capacity, gigawatts	340	530	1000
New installations, gigawatts/yr	30	40	68
Base load—coal, oil, gas	10.1	12.7	5.2
Intermediate load—all fossil	6.2	13.3	23.4
Peaking load—all fossil	5.8	2.3	7.2
Electric utility power generation			
	1960	1968	
Coal, %	66.3	61.9	
Gas, %	26.0	27.6	
Oil, %	7.6	9.4	
Nuclear, %	0.1	1.1	
Installed costs of a coal-fired utility steam generator			
Generator and support, per kW	$35		
Generator and support (including draft feedwater, control, coal and ash handling, dust collection, piping, and other auxiliaries), per kW	$60		

3. Peaking load plants—with a load factor of less than 20 percent and operating with gas turbines.

The most obvious market for fluidized-bed combustion boilers are those now predicted for the base and intermediate load fossil-fired plants; this market is expected to increase by 70 percent—from 16 gigawatts in 1970 to 28 gigawatts in 1985. If the fluidized-bed boilers are effective in air pollution control and if their capital and operating costs are sufficiently low, they may capture an additional share of the generation market now ceded to nuclear-fired base-load plants and to oil-fired intermediate-load plants. Higher plant efficiencies might be possible with increased steam temperature and with pressurized boilers with combined gas/steam turbine cycle.

A gigawatt of power generating capacity is roughly equivalent to 10×10^6 lb/hr of steam generating capacity and a coal consumption rate of 400 tons/hr. The present utility boiler market is therefore larger than the industrial boiler market by a factor of 2 in terms of capacity and by a factor of 7 in terms of money. Utility boilers also present a more serious problem in air pollution abatement. Purchasers of the smaller industrial boilers are turning to natural gas and low sulfur oil fuels. Because of coal's low cost and availability (despite recent problems of mining it in sufficient quantities), coal remains a prime fuel for the utilities. Most of the coal available to power plants in the eastern United States has quantities of sulfur beyond air pollution limits. A 1000-megawatt (1-gigawatt) plant burning 3.0-percent sulfur fuel products 24 tons/hr of SO_2. A fluidized-bed utility boiler, which can economically absorb this pollutant (permitting the recovery of sulfur) while minimizing the production of NO_x and particulates, can be of great benefit in reducing air pollution since about half of the SO_2 emissions and a quarter of the NO_x emissions are attributed to electric power generation by the utilities.

Functional Boiler Specifications

In addition to examining the past and future demand for industrial and utility boilers, the market survey has also considered customer requirements for the boilers they purchase. Figures 1 through 5 show capacity and steam conditions for future industrial and utility boiler markets.

Figure 1 shows that the largest portion of the industrial boiler market will be supplied by units in the capacity range of 150,000-250,000 lb/hr, a size that, not coincidently, is the largest that can now be factory assembled and shipped as a package. If a fluidized-bed boiler can be more compact than a packaged gas- or oil-fired boiler of conventional design, larger industrial boilers may become more popular.

Figures 2 and 3 show that the steam conditions of pressure and temperature in general exhibit a broad range of preference: about two-thirds of the boilers have steam pressures below 600 psi and temperatures below 750°F.

Figure 4 shows that the largest share of the fossil-fired utility boiler market will be supplied by units in the 400-600 megawatt range; a number of waste-heat recovery boilers in the 100-200 megawatt range may also be purchased to operate in conjunction with gas turbines in combined cycle power plants. Figure 5 shows that super-critical boiler units operating at around 3600 psi and sub-critical units operating at around 2400 psi steam pressure will continue to be popular with customers.

Table 3 summarizes the information on functional requirements used in establishing specifications for the industrial and utility fluidized-bed boiler designs. The final designation of specifications is discussed in papers dealing with the detailed boiler designs.

Fossil Fuels for Boilers

Table 4 shows the proved recoverable fuel reserves available for possible use in the U.S. boilers. Actual economically recoverable reserves of these fossil fuels may be as much as 2-3 times the proved values; but still the message appears clear. Coal is the most plentiful source of fossil energy. Domestic oil and gas reserves are in relatively short supply. Their prices can be expected to increase especially if foreign developments hinder the free flow of oil to this country. Slightly over half of the coal energy of the U.S. is in deposits east of the Mississippi River: most of this coal has a sulfur content of 2.5-4.0 percent by weight. Ultimately industrial and utility boiler operators will probably be required to utilize coal or a coal-derived fuel.

Table 5 and Figure 6 summarize predicted costs of fossil fuels. Major factors–including production costs, alternate market opportunities, and government regulation–influencing the market price of each are different. Political and environmental constraints, already shaping the price, supply, and demand of each fuel, will continue to affect fuel price trends over the next 15 years.

Table 6 gives transportation costs for fuels. Costs indicate that coal and gas cannot be used economically in large utility boilers far from the source of supply.

Table 7, based on a careful consideration of all the technical, economic, and political factors involved in the choice of fuel supply, indicates that utilities will use fossil fuels for generating over two-thirds of the electric

Table 3. FUNCTIONAL SPECIFICATIONS FOR FLUIDIZED-BED COMBUSTION BOILERS DEVELOPED BY THE BOILER MARKET SURVEY

Characteristic	Industrial boiler	Utility boiler		
Capacity				
Maximum	350-500 x 10^3 lb/hr	1300 MW		
Most frequent	150-250 x 10^3 lb/hr	500-700 MW		
Minimum	25 x 10^3 lb/hr	100 MW		
Steam conditions				
	Size, MW	<300	500-800	<800
Pressure, psig	150-600	1800	2400	3600
Temperature, °F	(sat.–775)	950-950	1000-1000	1050-1050
Performance				
Efficiency, %	86	89-92		
Special requirements				
Turndown	1/3	1/4 (1/2)		
Overpressure & flow %	–	5 (on each)		
Dynamics, %/minute	–	5		
Startup, shutdown	–	Automatic		
Pressurized operation	No	Yes		
TV viewing	No	Yes		
Packaged construction	Preferred	Unfeasible at present, but desired		
Multifuel capability	Not usually demanded	Desired		

power in 1980. A similar prediction for industrial boilers is much more difficult at this time. In any case, the development of an adequate solution to the air pollution abatement problem is a very important factor in determining the choice of fuels and in the economics of steam and power generation.

Table 4. PROVED RECOVERABLE RESERVES OF FOSSIL FUELS IN 1968

Fuel	Proven reserves	Annual consumption
Coal, U.S., tons x 10^9	265	0.5
Gas, U.S., MCF x 10^9	287	19.9
Oil, U.S., bbls x 10^9		
U.S.	31	4.78
Balance free world	381	7.57
Communist block	59	2.08
Alaskan field (est)	20-30	––

Alternate Means of SO_2 Pollution Control in Boilers

A number of stack-gas processing systems have been proposed for SO_2 removal. These systems have been considered for possible use with conventional coal-fired boilers in electric power plants. Some of these systems are ready for commercial application or nearly so; some require additional development and testing. Table 8 estimates their effectiveness and cost.

In general, the capital costs of the systems are from a third to half the cost of the utility steam generator itself. Monsanto's catalytic oxidation costs equal the \$35 per kilowatt cost of the boiler; TVA's dry lime process is about a quarter of this amount. While most of the processes claim the 90-percent SO_2 removal required by today's air pollution abatement goals, it is not clear how many of

Table 5. PRICE TRENDS OF FOSSIL FUELS FOR POWER GENERATION

Fuel	1968 Price, ¢/10^6 Btu	Price Indices (1968 = 100) 1970	1975	1980	1985
Coal (mine mouth)					
Steam coal	17	218	205	243	291
Gas	25	108	287	347	386
Residual oil (sea terminal)					
Low-S (0.3%)	32	230	267	291	295
High-S (2-1/2%)	32	144	141	148	156

Table 6. COMPARATIVE TRANSPORTATION COSTS FOSSIL FUELS FOR ELECTRIC UTILITIES

Fuel	Cost[a]
Coal (unit train)	
Under 300 miles	3.34
Over 600 miles	2.92
Gas (48-in. pipeline)	1.50
Residual oil	
Large tankers	0.40
Large river barges	0.60

[a] Cost in ¢/million Btu/100 miles.

them can economically accomplish the 95-98 percent removal which may ultimately be required as the use of coal in power generation increases. These stack-gas cleanup processes are unlikely to be economic for industrial boilers. Scaled down to a size corresponding with 250,000 lb/hr steam boiler, their cost would be in the range of $1.50-$3.00 per lb/hr. greater than the cost of the boiler itself.

If fluidized-bed combustion boilers are more effective in air pollution control and lower in cost than conventional boilers plus stack-gas cleanup equipment, they may capture a sizeable portion of the predicted market.

Pollution Control Targets

Projections of power generation by fossil fuels have been used to estimate annual emissions of SO_2, NO_x and particulates to the year 2000. These estimates are based on the following assumptions:

1. Sulfur content of coal increases from the current 2.7 percent to 3.5 percent in the year 2000.
2. Coal and oil generation are combined.
3. Nitrogen oxides are produced at a rate of 800 lb/billion Btu for coal and oil.
4. Heat rates are 10,300 Btu/kWh for coal and oil and 12,500 Btu/lb for coal.
5. Flue-gas/coal ratio by weight is 12.5.

Tables 9, 10, and 11 show the degree of pollutant reduction or removal required to achieve acceptable air standards in 1970 and to maintain the same total emissions (despite increasing quantities of fossil fuels used in power generation) in 1985 and 2000. Although 90-98 percent reductions of SO_2, NO_x, and particulates may be beyond the state of present art, such reductions are the targets of fluidized-bed boilers. As such, they provide a basis for design and a direction for future development.

Table 7. RELATIVE SHARE OF ELECTRIC UTILITY STEAM GENERATION

Fuel	1969		1970		1975		1980		1985	
	kWh x 10^9	%	kWh x 10^9	%	kWh x 10^9	%	kWh x 10^9	%	kWh x 10^9	%
Fossil	1283.0	98.5	1366.0	95.6	1741.0	79.1	2028.0	68.5	2416	59.5
Coal	793.1	60.9	858.8	60.1	1101.7	50.0	1252.5	42.3	1396.1	34.4
Gas	354.6	27.2	371.5	26.0	461.7	21.0	536.0	18.1	625.4	15.4
Oil	135.3	10.4	135.7	9.5	177.6	8.1	239.5	8.1	394.5	9.7
Nuclear	19	1.5	63	4.4	461	20.9	933	31.5	1648	40.5
Total	1302	100	1429	100	2202	200	2961	100	4064	100

Table 8. ASSESSMENT OF FLUE GAS DESULFURIZATION PROCESSES

Process	Plant size, MW	Process cost Capital $/kW	Process cost Operating ¢/10^6 Btu	Years to full-scale plant	% SO_2 removal	Chance of success
Ready for commercial application						
C.E. dry lime-wet scrub	500	11	4.2	3 sold	85	Good
Monsanto cat-ox	500	36	0.2[a]	ready	90	Good
Nearly ready for application						
NAPCA-TVA dry lime	500	8	4.3	ready	40	Fair
Wellman-Lord	500	20[b]	3.9[a,b]	1 year	90	Fair
In development						
S & W-Ionics electrolysis	1200	19[b]	0.6[a,b]	5 years	90	Fair
Esso-B&W dry adsorb	800	17[b]	0 [a,b]	6 years		Unpredict
A.I. molten carbonate	800	15[b]	4 [a,b]	7 years		Unpredict
Consol Coal potassium formate	1300	17[b]	5 [a,b]	7 years	90	Unpredict

[a] Assumes byproduct credit but no credit for lower stack, eliminated precipitator, etc.

[b] Optimistically low cost, estimated by process developers.

Table 9. PROJECTED SO_2 EMISSION LEVELS FROM POWER GENERATION PLANTS

Year	Control level[a]	Average SO_2 leaving stack, ppm	SO_2 reduction, %
1970	I	2000	0
	II	2000	0
	III	220	89
1985	I	2220	0
	II	1230	45
	III	140	94
2000	I	2600	0
	II	500	81
	III	55	98

[a] Control levels are: I—uncontrolled; II—equivalent to maintaining total SO_2 emissions from power plants at the 1970 level; and III—equivalent to SO_2 emissions if all the 1970 generating capacity was based on using ~0.3 percent sulfur fuel.

Table 10. PROJECTED NITROGEN OXIDES EMISSIONS FROM POWER GENERATION PLANTS

Year	Control level[a]	Average NO_x leaving stack, ppm	NO_x reduction, %
1970	I	520	0
	II	520	0
	III	260	50
1985	I	520	0
	II	320	39
	III	160	69
2000	I	520	0
	II	130	75
	III	65	88

[a] Control levels are: I—uncontrolled; II—equivalent to maintaining total emissions to the atmosphere from power plants at the 1970 level; and III—equivalent to reducing the total NO_x emissions to 50 percent of the 1970 level.

BOILER DESIGN

The results of the boiler market studies and the air pollution abatement considerations have been used in establishing functional specifications for one industrial and two utility boilers. A number of boiler concepts have been reviewed and evaluated to determine their compatibility with these specifications and the general and economic requirements of the market.

Functional Specifications for a Fluidized-Bed Industrial Boiler

Results of these considerations, with regard to the utility boiler, are presented in a companion paper. This paper deals with the industrial boiler. Table 12 gives its functional specifications. A 250,000-lb/hr capacity was chosen because of the market for this size and because it may be the largest fluidized-bed boiler shippable as a single module. A steam pressure of 600 psi with superheat from the saturation temperature of 489°F to 750°F has been selected; these values have been chosen because two-thirds of the industrial boilers operate at or below these conditions. The fuel for this boiler is a crushed Pittsburgh coal, undried, with 4.3 percent sulfur. (Pulverized coal has been ruled out because of the expense and operating problems involved with coal mills.)

Table 11. PROJECTED PARTICULATE EMISSIONS FROM POWER GENERATION PLANTS

Year	Control level[a]	Emission, gr/SCF	Reduction, %
1970	I	3.7	0
	II	0.5	87
	III	0.037	99
1985	I	3.7	0
	II	0.30	92
	III	0.023	99.4
2000	I	3.7	0
	II	0.12	97
	III	0.009	99.8

[a] Control levels are: I–uncontrolled; II–equivalent to maintaining total emissions rfom power plants at 1970 control level of ∿ 87-percent removal;[b] and III–equivalent to maintaining total emissions from power plants at a level corresponding to 99-percent removal in 1970.

[b] NAPCA. Control Techniques for Particulate Air Pollutants. Publication No. AP-51. Washington, D. C., January 1969.

Reduction of SO_2 emissions by 90 percent is to be accomplished by lime (CaO, $CaCO_3$, or dolomite) additions to the bed. Alternate

Table 12. FUNCTIONAL SPECIFICATIONS FOR A FLUIDIZED-BED INDUSTRIAL BOILER

Characteristic	Specification
Steam capacity, lb/hr	250,000
Steam conditions	
Pressure, psig	600
Saturation temperature, °F	489
Superheated to, °F	750
Water to economizer at, °F	250
Water leaving economizer at, °F	350
Fuel	
Coal	Ohio Pittsburgh No. 8 seam
Crushed to	-1/4 in. x 0
Air pollution control targets	
SO_2 reduction by limestone absorption, %	90
NO_x	
Partciulates reduction, %	95[a]
Boiler efficiency, %	>85
Packaged construction	
Factory fabrication	Yes
Shippable	Yes
Dimensional limits of primary modules, ft	12 x 16 x 40

[a] All ash and 3 percent of lime assumed to be elutriated.

absorbents are not considered because their cost is in general higher and their effectiveness is not yet demonstrated. A regeneration system is specified for the lime absorbent because large quantities of this material will probably be required to achieve a 90-percent reduction in SO_2 emissions. It does not seem economically or technically feasible to dispose of large amounts (up to half the coal tonnage) of spent absorbent. Regeneration with sulfur recovery does not appear easy or cheap but it remains to be evaluated. A more economic solution for industrial applications may be to use a desulfurized coal char in a fluidized-bed boiler. Such a boiler is probably the only economic means for using char while minimizing NO_x and particulate emissions.

Reduction of the particulate ash and limestone emissions in the stack gases is to be achieved by a combination of cyclones and, if required, an electrostatic precipitator.

Finally, an overall boiler efficiency greater than 85 percent has been specified in order to obtain reasonable fuel costs.

Fluidized-Bed Industrial Boiler, Operating Conditions

Figure 7 shows that the fluidized-bed boiler comprises three main sections–the combustor and SO_2 absorber, the carbon burnup cell (CBC), and the lime regenerator. The functions of the last two sections might be combined if the amount of carbon elutriated from the combustor can be minimized. When large amounts of carbon carryover must be consumed, combining the CBC and the regenerator produces a gas mixture dilute in SO_2. This bed can be operated intermittently, varying temperature and flue gas composition, to produce periodic surges of SO_2 from the combined CBC/regenerator. But in either case, capture or recovery of the sulfur is complicated.

About 85 percent (11 ton/hr) of the total coal required is fed into the primary fluidized-bed combustor (FBC) with coal combustion efficiency of 87 percent. The remaining 13 percent unburned coal is elutriated, along with the ash and 3 percent of the spent limestone. Ninety-five percent of the elutriated solids from the FBC is recycled through the primary cyclone back to the CBC for further combustion. In addition to the recycled coal, fresh coal (about 15 percent of total coal required) is also fed to the CBC. Consequently, about 75 percent of the total coal is burned in the FBC; the other 25 percent is burned in the CBC. The ash and spent limestone recycled back to the CBC are again elutriated along with additional ash and spent limestone produced in the CBC and eventually collected in a secondary cyclone and electrostatic precipitator.

The spent limestone from the combustor is regenerated in the limestone regenerator. The total SO_2 removed in the regenerator is assumed to be 90 percent of that picked up in the combustor. The average composition of the recycled limestone is assumed to have 5 mole percent sulfate. To maintain reactivity, 10 percent of the recycled limestone is rejected and replaced by fresh calcined lime.

Table 13 details operating conditions for the boiler. Bed temperatures chosen are based on the experience of all those in the NAPCA program: they are high enough to obtain good combustion and heat transfer, yet low enough to obtain good performance from the lime absorbent. High gas velocities (15 ft/sec) in the beds have been specified to achieve high boiler compaction. A large top size of the crushed coal (1/4 in.) has also been specified to minimize elutriation and to ease coal distribution problems: fine particles tend to devolatilize and burn closer to the point of their introduction. Coarser particles are expected to burn more uniformly throughout the bed.

Minimizing elutriation has also been an important consideration in choosing a particle size distribution for the lime absorbent. A

Table 13. OPERATING CONDITIONS FOR A FLUIDIZED-BED INDUSTRIAL BOILER

Condition	Fluidized-bed combustor	Carbon burnup cell	Limestone regenerator	Boiler
Temperature, °F	1650	1900	2000 (bed)	–
Coal combustion efficiency, %	87	90	–	–
Air excess, %	10	30	–	–
Superficial fluidizing velocity, ft/sec	15	15	–	–
SO_2 removal, %	–	–	90	–
Regenerating gas	–	–	$CO_2/CO = 2$	–
Particulate removal efficiency, %	–	–	–	95
Stack gas temperature, °F	–	–	–	350
Air preheat temperature, °F	–	–	–	None
Boiler efficiency, %	–	–	–	86.2
Heat losses				
Dry flue gas, %				5.65
Evaporation of water formed by burning hydrogen, %				4.37
Evaporation of water in fuel, %				0.30
Heating water in air, %				0.14
CO and unburned C, %				3.00
Radiation, %				0.36

Limestone feed

Stone–BCR 1359 as the primary bed material with characteristics

Particle size–1000-5000 microns with average diameter ∿2500 microns

Feed requirements–∿90% of SO_2 removal with 6 times stoichiometric feed ratio

SO_2/CaO reaction heat generation–3×10^6 Btu/ton $CaSO_4$ produced and the stone enters boiler at CaO at 1900°F after regeneration

Recycle rate–3% elutriated with the fly ash and 10% rejection in the regeneration

lime flow rate has been specified to yield a molar Ca/S ratio of 6. Data from NAPCA contractors indicates that this ratio should be adequate to obtain a 90 percent reduction in SO_2.

Excess air amounting to 10 percent is fed to the combustor; and 30 percent to the CBC. The regenerator is operated with an air deficiency so that the molar CO_2/CO in the SO_2 containing flue gases is 2.

The temperature of the stack gases leaving the boiler has been specified as 350°F; water temperatures entering and leaving the economizer, as 250 and 350°F. These temperature designations are based on conventional boiler practice.

Preliminary Design Computations and Considerations

Material and heat balances, together with heat transfer computations, have been carried out for the fluidized-bed industrial boiler; some of the results of this work are presented in Table 14 and Figures 7 through 11. Figure 7 shows the large quantities of lime absorbent and particulate solids which must be handled in the system; it seems important to closely integrate both the lime regenerator and CBC with the fluidized-bed combustor and sulfur absorber. This integration can minimize the distances over which solids must be transported.

Figures 8 through 11 are heat balances, in the form of temperature-enthalpy diagrams, for various fluidized-bed boiler functional arrangements. These diagrams show temperatures throughout the boiler and the corresponding heat quantities transferred from the burning solids or combustion gases to the water and/or steam expressed in terms of Btu per lb of fuel burned. About 20 percent of the heat released increases the water temperature from 250°F to the saturation value.

Table 14. HEAT TRANSFER COMPUTATIONS FOR FLUIDIZED-BED BOILER DESIGNS

Characteristic	Shop-assembled conventional boiler	FBC boiler	
		Vertical tubes in bed extending into freeboard	Horizontal tubes in bed
Heat load, Btu/hr x 10^6			
Submerged in bed	78.0	136.8	136.8
Above bed		63.4	10.8
Superheater	26.1	43.6	43.6
Convection pass	143.4	20.8	73.3
Total	247.5	264.6	264.5
Log mean temperature difference, °F			
Submerged in bed	1910	1161	1161
Above bed		742	1075
Superheater	1410	1280	1280
Convection pass	796	291	495
Heat transfer coefficients, Btu/ft^2/hr/°F			
Submerged in bed	29.2	50	50
Above bed		30	30
Superheater	30.4	30	30
Convention pass	20.9	20.4	30
Heat transfer surface, ft^2			
Submerged in bed	1400	2350	2350
Above bed		2840	335
Superheater	609	1140	1135
Convection pass	8630	3510	4950
Total	10639	9840	8770

489°F; 50 percent vaporizes the water; and 15 percent superheats the steam to 750°F.

Figure 8 corresponds to a fluidized-bed boiler with vertical evaporator tubes extending through the bed and freeboard. (These tubes might be distributed more or less uniformly or clustered in banks or platens.) The tube surface above the bed cools the gases from the 1650°F bed temperature to 925°F. A convection pass further cools the gases to 672°F; and an economizer recovers the balance of the heat from the stack gases. The superheater is located in the CBC; sufficient coal is fired with the char to provide the necessary heat. Dotted lines show a possible alternate of bringing water to saturation in the bed and using the convection pass as an evaporator.

Figure 9 corresponds to a boiler with horizontal tubes in the fluidized beds. In this particular situation, the CBC does not superheat all of the steam; a portion (24 percent) of the heat released by combustion gases as they are cooled from 1650 to 1530°F is transferred to the steam in a convection superheater.

Figures 10 and 11 show two of many alternate arrangements of different functional units of an industrial boiler. In Figure 10 an air preheater is used which, in this example, transfers the same amount of energy that the economizer does and preheats the air from 80 to 430°F. For each Btu of heat transferred to the combustion air in a preheater, an additional Btu must be transferred to the steam in the fluidized-bed combustor if the bed temperature is to remain constant. The economizer is put into the fluidized bed, and the convection pass surface is packed above the fluidized bed to take advantage of higher heat transfer coefficients. The economizer is eliminated in Figure 11 and the heat transfer surface above the bed preheats the water. The superheater is put in the CBC with a small portion in the fluidized bed of the main carbon combustor; in another alternative, the superheater is in the fluidized-bed combustor and the CBC serves as an evaporator. Numerous other arrangements can also be proposed; the optimum design will depend not only on the economic consideration but also on steam quality, velocity in the tubes, and the turndown characteristics of the boiler plant. Consideration of all the possible alternatives is a way to an optimum design.

Preliminary heat transfer computations have been carried out on the vertical and horizontal tube designs whose heat balances are represented in Figures 8 and 9. Table 14 compares the results of these computations with similar results for a conventional oil-fired packaged boiler. The fluidized-bed boilers transfer 7 percent more heat and have 10 percent less transfer surface. Heat transfer coefficients above the bed and in the superheater have been chosen conservatively low (30 Btu/ft^2/hr/°F); they may well be 40 Btu/ft^2/hr/°F or higher. In this instance, a fluidized coal-fired boiler will have 30 percent less heat transfer surface than a comparable convection gas- or oil-fired boiler.

For NAPCA, and under subcontract to Westinghouse, Erie City is providing proposal designs of an industrial boiler whose functional specifications, operating conditions, and material and heat balances have been discussed. Erie City will describe its work in a companion paper.

PROBLEMS IN BOILER DESIGN AND DEVELOPMENT

Proposal designs of one industrial and two fluidized-bed boilers are now being prepared. The primary purpose of these designs is to evaluate the effectiveness and economics of such boilers in generating steam and power without air pollution. Their secondary purpose is to reveal areas in which additional knowledge or more development is needed to produce a commercial fluidized-bed boiler.

Although designs are still far from complete, some problem areas have already been encountered. A companion paper discusses those involved with a pressurized utility boiler; this paper is concerned with the problems involved with the design of an atmospheric industrial boiler. These problems fall into two areas–those involved primarily with operation and those with design.

Operational Problems

The most important choice in determining the mode of operation of a fluidized-bed boiler is that of gas velocity (and of the corresponding bed particle size). A high gas velocity, together with a coarse coal and limestone, has been chosen for the industrial boiler to minimize the cross-sectional bed area. Hopefully the boiler will then be most compact. Although coarse coal may also minimize coal distribution problems, coarse particles result in lower heat transfer coefficients. High air velocities result in: greater elutriation of carbon from the bed; higher attrition in the limestone absorbent; and, perhaps, erosion of the heat transfer surfaces. The problem is to determine an optimum gas velocity-particle size choice; present performance and economic data do not appear adequate to make this choice.

A second operational problem is that of boiler turndown; several procedures are possible:

1. "Slumping" the bed; i.e., merely decreasing the air flow and depending on bed cooling, bed contraction, or defluidization to reduce heat transfer and thus steam generation.
2. Decreasing air flow and simultaneously draining a portion of the solids from the bed to reduce heat transfer area.
3. Shutting off the air flow to whole sections of the boiler.

Selecting the procedure that will provide the greatest flexibility in turndown, yet minimize operating problems and boiler costs, is a problem.

Design Problems

The most important design problem is designating the heat transfer surface arrangement. Vertical tubes permit natural circulation, but horizontal tubes with forced circulation may be more compact. The choice of tube orientation affects the air distributor, coal feeding, and tube manifolding systems. Also the choice of tube diameter and pitch has considerable effect on the design and operation of the boiler. Small tube diameters and close spacing produces a compact boiler; however, they may also result in poor fluidization and uneven fuel distribution. Tube erosion, clinkering, and excessive carryover of carbon may also occur.

A second design problem is the choice and location of the various boiler functions–air preheater, economizer, evaporator, and superheater–in the boiler. It is not clear than an economizer alone is the most effective and economic heat trap; it may be better to combine an economizer and an air preheater. Other questions involve the location of evaporators and superheaters in the beds and in the convection passes: Which should be placed where? And should the superheater be distributed throughout the boiler to facilitate turndown by sections?

A third design problem concerns using horizontal baffle tubes at the surface of the fluidized bed to minimize the splashing and spouting of particles. Baffle tubes are effective in minimizing the carryover of particles. And heat transfer coefficients for these tubes are apparently almost as high as for submerged tubes. But the reduction of particles above the bed may reduce heat transfer rates in the freeboard above the bed surface. Too severe cooling of the bed gases may inhibit combustion processes in the freeboard and result in increased carbon monoxide and hydrocarbon losses. Data is lacking for the design of an optimum baffle tube and freeboard section of a fluidized-bed boiler.

Other Problems

Two other general problems encountered in the design of a fluidized-bed industrial boiler have already been mentioned.

1. Designation of an optimum sulfur cleanup system. Lime has been designated as the absorbent, but should it be a once-through or a regenerative process? How should byproduct sulfur be recovered? Might it be more realistic in the long run to use a desulfurized char fuel?
2. Designation of an optimum design and operating conditions for NO_x and particulate emissions control. Data on simultaneous control of SO_2, NO_x, and particulates is inadequate to choose an optimum method. Might a two-stage combustion process be used to minimize NO_x formation? Can an electrostatic precipitator be used effectively to remove fly ash from fluidized beds since carbon may provide some electrical conductivity of the carryover?

CONCLUSIONS

Market studies for industrial and utility boilers, together with studies of fuel availability and of stack gas cleaning processes, indicate that a coal-burning, sulfur-absorbing fluidized-bed boiler can have a significant effect on both air pollution abatement and on the economic generation of steam and power. Functional specifications, including target values for SO_2, NO_x, and particulate reductions, have been drawn up for an industrial boiler. Operating conditions have been suggested, preliminary engineering computations have been performed, and proposal designs and evaluations are underway. Some problem areas require further data and development to confirm the practicality and effectiveness of fluidized-bed boilers.

ACKNOWLEDGEMENT

The work discussed in this paper was carried out under the sponsorship of the National Air Pollution Control Administration, Department of Health, Education, and Welfare. Mr. P. P. Turner monitored the work for NAPCA.

The authors wish to thank for their cooperation and support: Messrs. H. L. Smith and S. J. Jack for their work on the analysis and projections of the utility boiler market; Erie City Energy Division of Zurn Industries (Messrs. R. V. Seibel and W. D. Schwinden) for their work, under contract to Westinghouse, on industrial boiler market analyses and industrial fluidized-bed boiler design; and the NAPCA contractors working on fluidized-bed combustion.

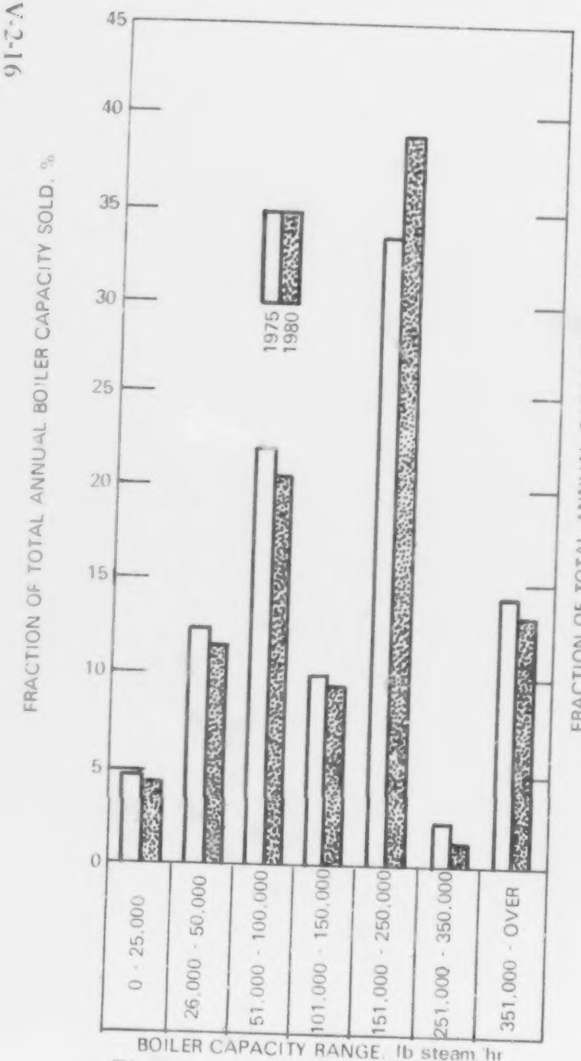

Figure 1. Projected distribution of U.S. industrial boiler sales -- steam generating capacity.

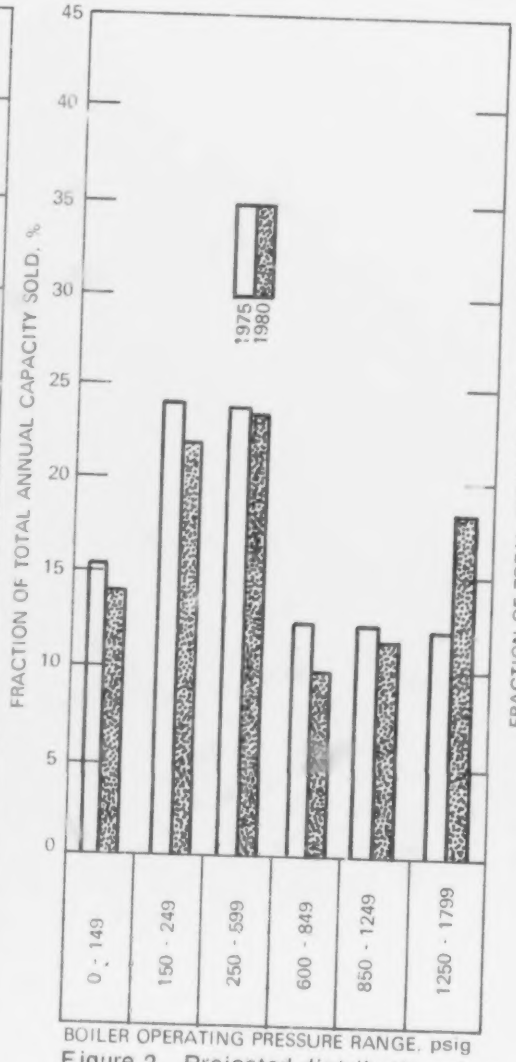

Figure 2. Projected distribution of U.S. industrial boiler sales -- operating pressure.

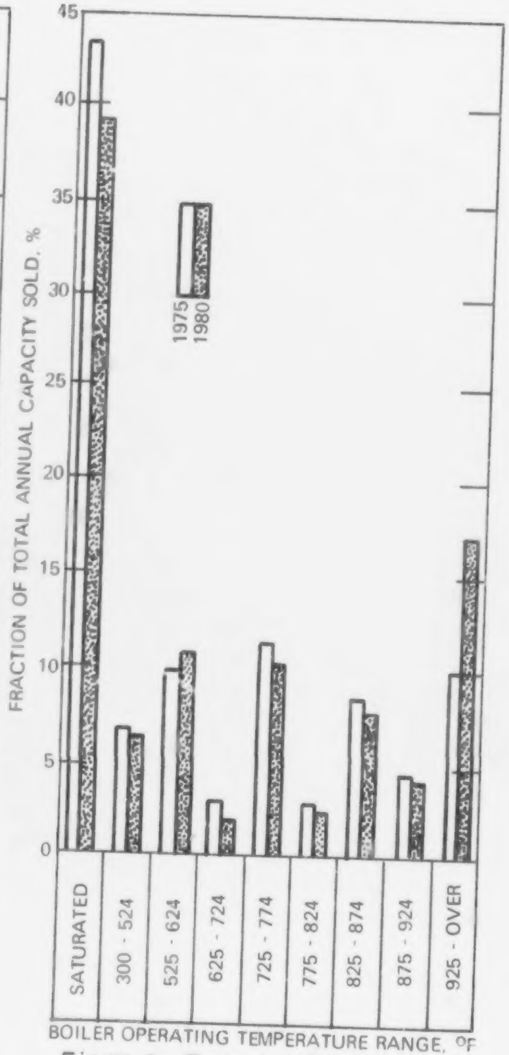

Figure 3. Projected distribution of U.S. industrial boiler sales -- operating temperature.

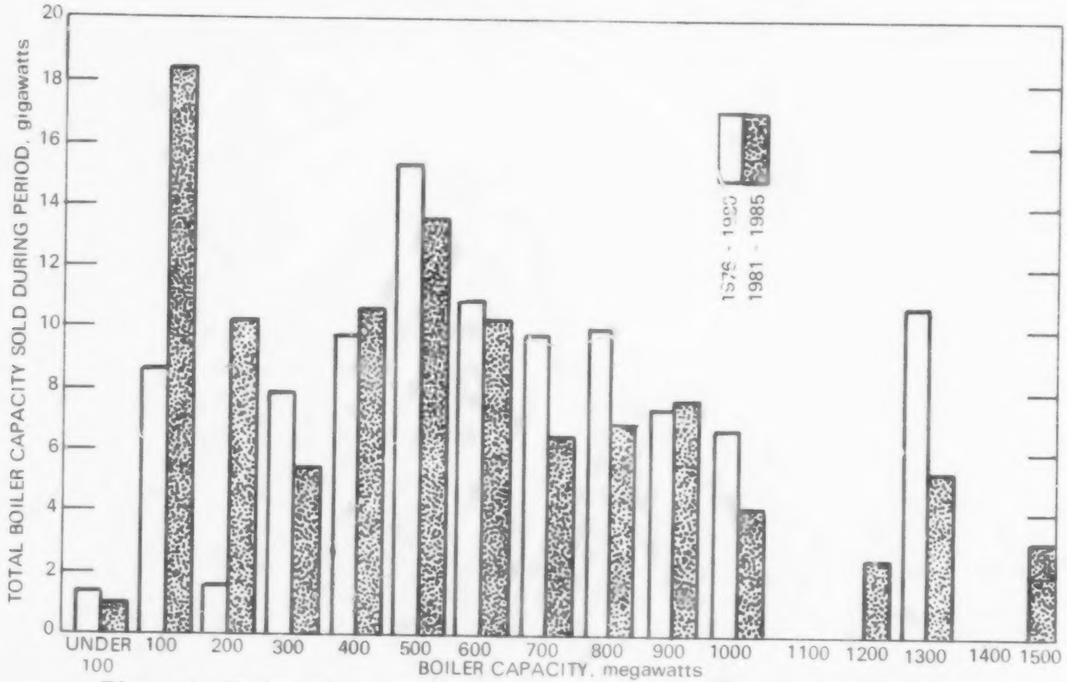

Figure 4. Projected U.S. utility boiler sales -- steam generating capacity.

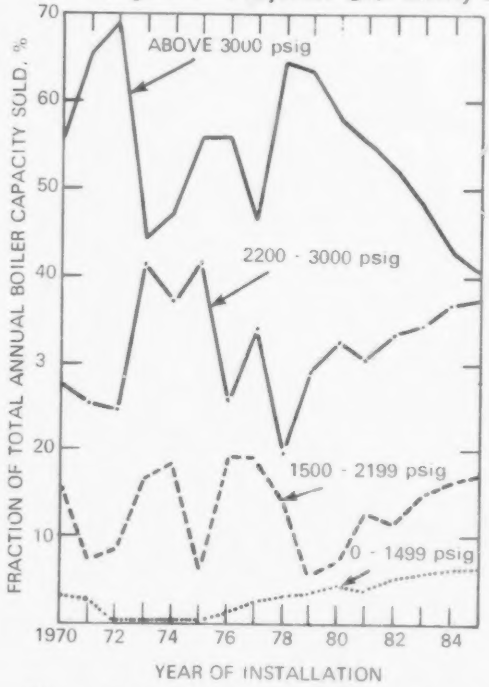

Figure 5. Projected distribution of U.S. utility boiler installations--steam pressure.

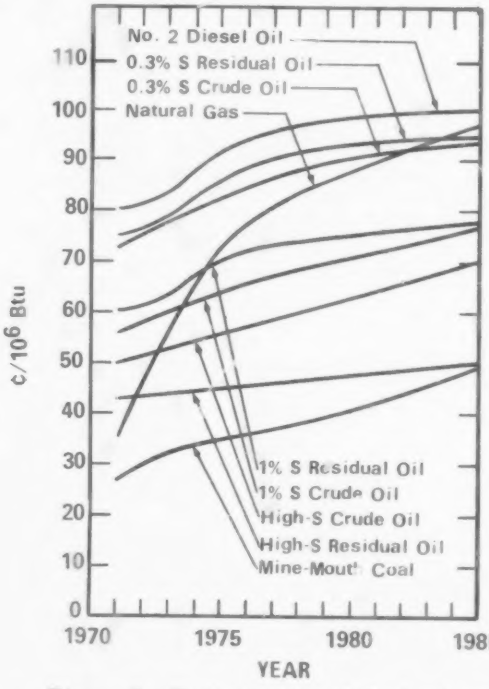

Figure 6. Projected fossil fuel prices for U.S. electric industry -- 1971-1985.

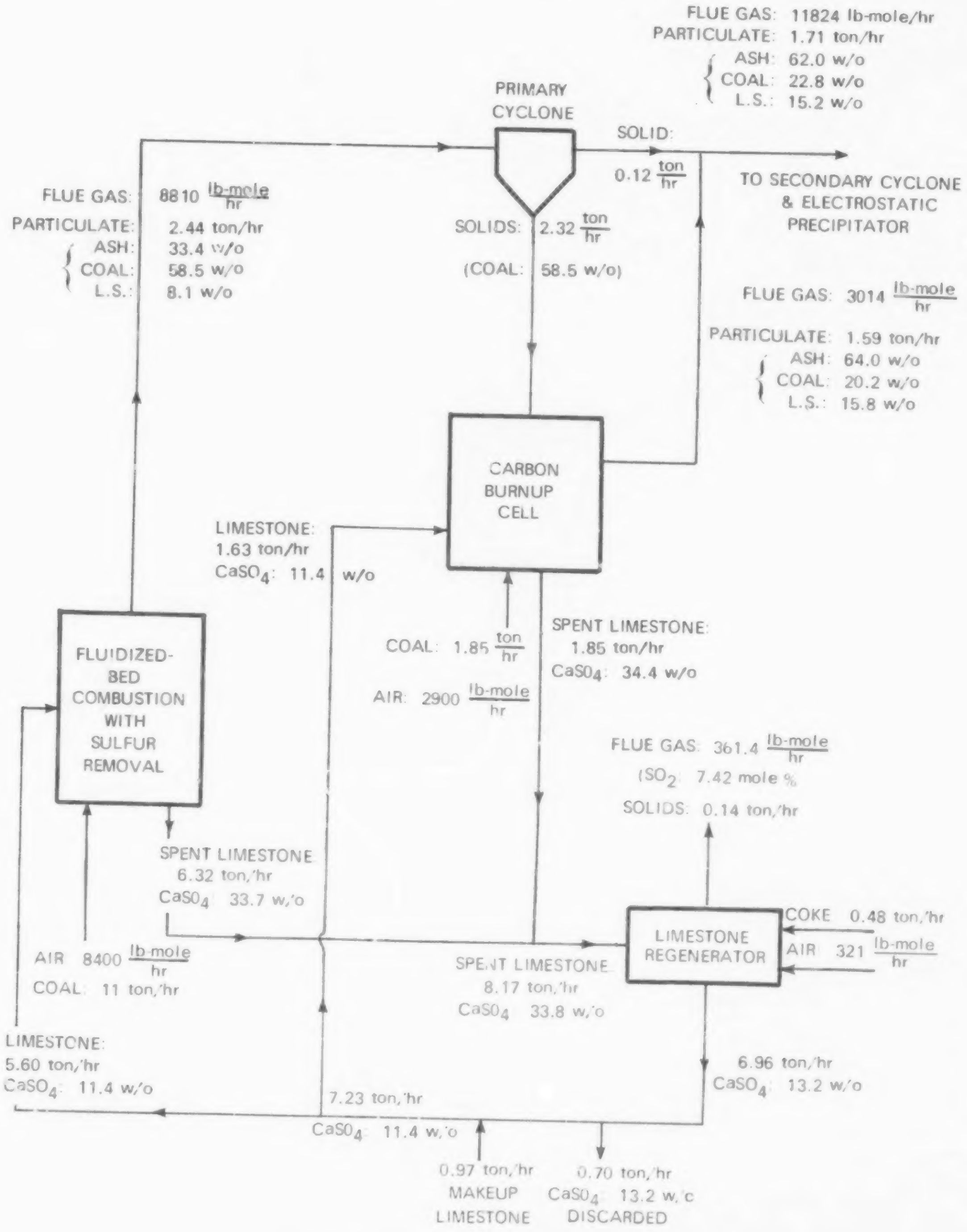
FLUE GAS: 11824 lb-mole/hr
PARTICULATE: 1.71 ton/hr
ASH: 62.0 w/o
COAL: 22.8 w/o
L.S.: 15.2 w/o
PRIMARY CYCLONE
SOLID: 0.12 ton/hr
TO SECONDARY CYCLONE & ELECTROSTATIC PRECIPITATOR
FLUE GAS: 8810 lb-mole/hr
PARTICULATE: 2.44 ton/hr
ASH: 33.4 w/o
COAL: 58.5 w/o
L.S.: 8.1 w/o
SOLIDS: 2.32 ton/hr
(COAL: 58.5 w/o)
FLUE GAS: 3014 lb-mole/hr
PARTICULATE: 1.59 ton/hr
ASH: 64.0 w/o
COAL: 20.2 w/o
L.S.: 15.8 w/o
CARBON BURNUP CELL
LIMESTONE: 1.63 ton/hr
CaSO4: 11.4 w/o
COAL: 1.85 ton/hr
AIR: 2900 lb-mole/hr
SPENT LIMESTONE: 1.85 ton/hr
CaSO4: 34.4 w/o
FLUIDIZED-BED COMBUSTION WITH SULFUR REMOVAL
FLUE GAS: 361.4 lb-mole/hr
(SO2: 7.42 mole %
SOLIDS: 0.14 ton/hr
SPENT LIMESTONE: 6.32 ton/hr
CaSO4: 33.7 w/o
COKE 0.48 ton/hr
AIR 321 lb-mole/hr
LIMESTONE REGENERATOR
AIR 8400 lb-mole/hr
COAL: 11 ton/hr
SPENT LIMESTONE: 8.17 ton/hr
CaSO4: 33.8 w/o
LIMESTONE: 5.60 ton/hr
CaSO4: 11.4 w/o
6.96 ton/hr
CaSO4: 13.2 w/o
7.23 ton/hr
CaSO4: 11.4 w/o
0.97 ton/hr MAKEUP LIMESTONE
0.70 ton/hr CaSO4: 13.2 w/o DISCARDED

Figure 7. Overall material balance.

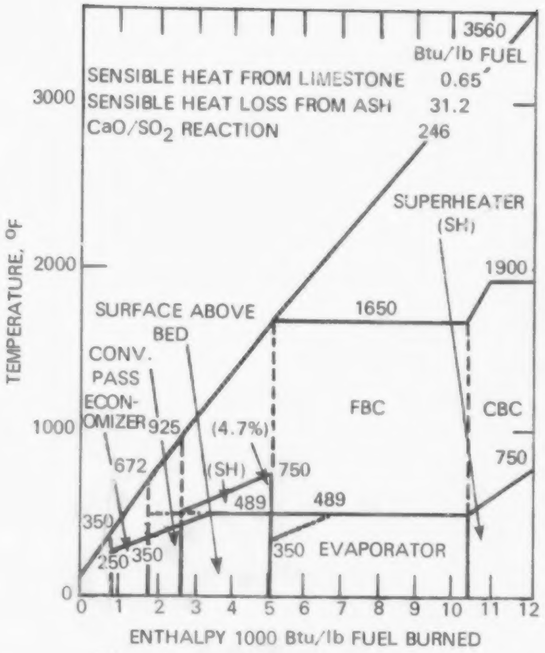

Figure 8. Temperature-enthalpy diagram for vertical bed tube design.

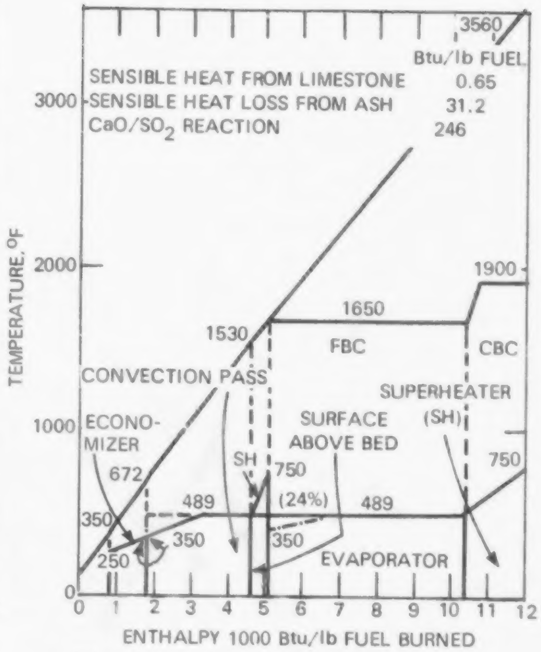

Figure 9. Temperature-enthalpy diagram for horizontal bed tube design.

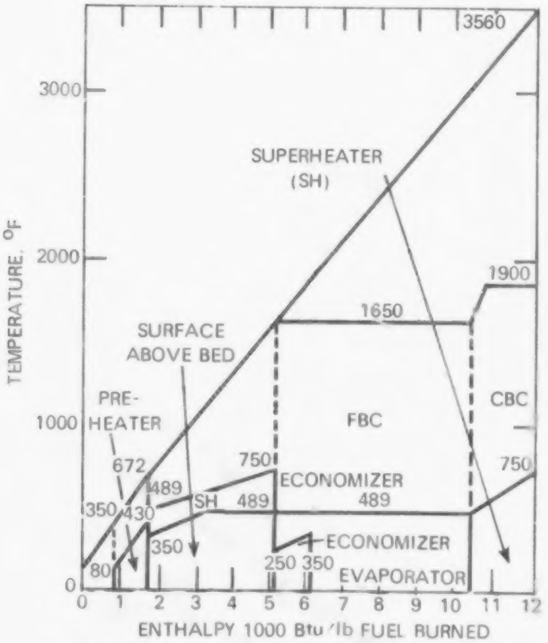

Figure 10. Arrangement of functional units of a fluidized-bed boiler -- alternative No. 1.

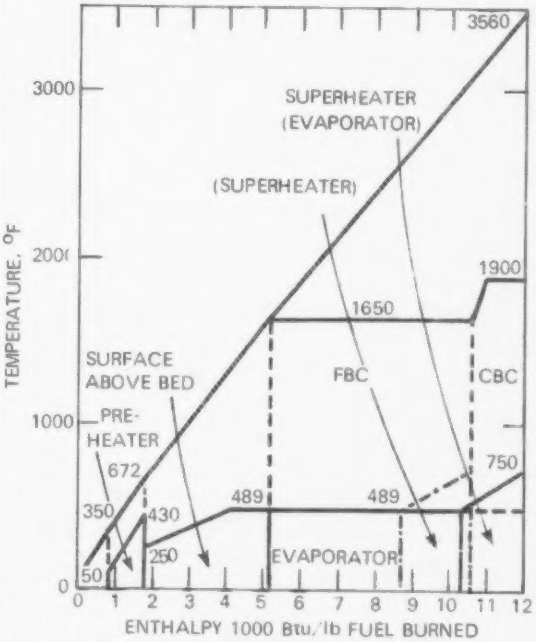

Figure 11. Arrangement of functional units of a fluidized-bed boiler -- alternative No. 2.

3. DESIGN OF ATMOSPHERIC FLUIDIZED-BED COMBUSTION STEAM GENERATOR

R. V. SEIBEL

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INTRODUCTION

If steam were an end product. studies of capital expenditures, operating costs. and maintenance costs for various steam-generator configurations would have precise meanings to all. Since steam is not an end product and a steam generator is never the only process in a plant, it follows that the worth of a steam generator to any particular user is a complex subject. In one case, it does not have the same meaning to all users.

Basically, a steam generator converts chemical energy (available from oxidizable organic materials) into heat energy (in a form that is transmittable to a remote process). Heat, in a convenient form, is the product of a steam generator. Producing heat for a process by converting chemical energy is the work of a steam generator. How much of the available chemical energy is converted? What procedure is best for making this conversion? How much does it cost to make this conversion? How much reliability is demanded in making the conversion? All these questions encompass the study of steam-generator design and application.

CONVENTIONAL STEAM GENERATORS

The four questions above (with the second and third combined for convenience) can be used as the basic thoughts in the study of fluidized-bed combustion techniques as applied to industrial applications in other terms in atmospheric design. This paper discusses basic facts and experience with conventional steam generators, and then summarizes initial work and thoughts on a fluidized-bed combustion (FBC) steam generator.

Quantity of Chemical Energy Converted

For all fuels, a terminal temperature of the combustion products (selected by experience) is the determining factor of boiler efficiency, defined as heat output divided by heat input. For natural gas, oil, and coal, the final or "stack" temperature for optimum boiler efficiency ranges from 300 to 350°F. Sensible and latent heat losses vary, depending on the fuel; at the same input and under identical conclusions: natural gas has the lowest efficiency; oil, the next; and coal, the highest.

Table 1 summarizes heat losses for three different fuels where the final combustion gas temperature is 300°F, and the sensible heat base is 80°F. Practical cases are shown where conventional design allows for varying excess combustion air and unburned combustible losses. No unburned combustible is generally found with the oxidization of natural gas and residual fuel oil. Practical designs of combustion methods of coal do not allow complete combustion of coal. The 3-percent combustible loss for coal in Table 1 represents spreader stoker firing for a bituminous coal.

Table 1 shows that boiler efficiency is greater for configurations of heat transfer surface on fuel burning equipment that allows lower final combustion product temperature, lower combustible losses, or reduced excess combustion air.

Process and Cost of Energy Conversion

Conventional steam generator designs promote rapid combustion reactions. The rapid combustion reactions produce very high temperatures; in fact, flame temperatures closely approach those produced at adabatic conditions, which are a factor in combustion chamber construction. It has become common practice to water-cool the combustion chamber to control maintenance from the high flame temperatures. Heat absorbed in the combustion chamber water-cooling system is recovered by generating steam.

In burning low ash fuels, the size of the combustion chamber is dependent on the fuel and the combustion equipment. A slow burning fuel or a low turbulence burner may dictate a larger residence time at high temperatures to complete combustion. Where ash in the fuel is molten at higher temperatures, the combustion furnace chamber water-cooling is used to lower (through radiant heat transfer) the combustion product gas temperature below the ash fusion temperature. In conventional design, flue gas temperatures leaving the combustion chamber range from 2000 to 2500°F.

Convection heat transfer is the principal way to reduce the furnace combustion products exit temperature to the final or stack temperature. Although convective heat

Table 1. COMPARISON OF HEAT LOSSES AND BOILER EFFICIENCY FOR THREE DIFFERENT FUELS[a]

Heat losses, % of heat input	Natural gas	Residual oil (#6)	Bituminous coal
Sensible heat in dry flue gas	4.2	4.5	5.0
Heat loss due to water in (and formed from) the fuel	10.6	6.0	4.2
Heat loss due to moisture in combustion air	0.1	0.1	0.1
Unburned combustible	–	–	3.0
Radiation and unaccounted for	1.5	1.5	1.5
Total heat losses	16.4	12.1	13.8
Heat output (boiler efficiency)	83.6	87.9	86.2

[a] Assumptions: combustion product final temperature–300°F;
natural gas excess combustion air–10%;
oil excess combustion air–15%;
coal excess combustion air–25%; and
type of coal firing–spreader stoker.

transfer depends somewhat on the local conditions of gas viscosity tube configuration and some other minor factors, it depends mostly on velocity. However, two factors limit obtaining very high coefficients.

One limit stems from conventional design practice which has been changing over the years in an attempt to optimize power and heat absorption rates for gas and oil fuels: power requirements for air-moving equipment are a direct exponential function of velocity. The other limit is the presence of particulate material in the flue gas, with erosion of construction materials a necessary consideration. For examples of the above, velocities in the convective zones for oil- and gas-fired steam generators have increased to 120 - 140 ft per second; for spreader stoker firing, velocities are normally limited to 75 ft per second. Convection heat transfer rates can be much greater for a particulate-free gas stream, or when the erosiveness of the particulates is less.

The temperature of flue gas leaving the steam generator (that is, upstream of the separate heat trap) is largely determined by practical design factors that are not apparent to the casual observer; optimization of supports, drum ligaments, fabrication capability, and assembly costs are just a few of the many factors that must be considered by the boiler designer. It is normal, however, to be able to achieve gas temperatures of about 700°F in industrial boiler practice. Useful heat remaining in the combustion product gas stream can be roughly computed from the above and corresponds to a 400°F temperature change. Often only one heat trap is necessary to produce the desired thermal efficiency of the steam generator system.

An important philosophy, here, is that the steam generator is a combination of combustion chamber and convection zone. Utilization of all the heating surface configuration that is physically possibly adds little more cost than only partial use. Heat traps, separate items, are an additional expense. An increase in heat duty assigned to a heat trap has a direct effect on the cost so that, if boiler heating surface is sacrificed to where heat traps must be used, overall costs must be increased. For example, if there is no convection surface in parallel with furnace tubes from the steam drum there is only partial utilization of the steam drum physical capabilities.

In the discussion of heat traps, there are two types of combustion air heaters and also flue gas contacting feedwater heaters or "economizers." There are regenerative and tubular air heaters; economizers may be "bare tube" or "extended surface." Where a combustion air heater is used, the heat absorbed is directed to the combustion chamber and convective boiler surface; where an economizer is used, there is no increase in heat input to the combustion chamber.

The coefficients that can be obtained and the heat transfer temperature differences favor the application of economizer, but a limiting factor is the amount of heat that can be absorbed in an economizer. Conventional design avoids obtaining saturated temperatures in the feedwater. Normal industrial applications have feedwater inlet temperatures of about 220°F, allowing a temperature rise of over 100°F in the economizer which is sufficient to achieve the desired boiler efficiency.

Combustion air heaters find a limit on the maximum possible air temperature as it affects the mechanical design. It is common, for example, to limit stoker combustion air temperatures to 400°F. Another limit is the "cold end" problem where metal parts in contact with flue gas are at the dew point. It is common to find a separate heater used to limit the lowest metal temperature to something above the flue gas dew point.

Operational Reliability

It is impossible for an unscheduled shutdown not to affect the economy of plant operation. Maintenance may be a trade off for operating economy or for lower capital cost, but designs that do not have predictable reliability are unacceptable.

It is very difficult to describe normally expected reliability. Reliability can vary from indefinite periods of operation approaching a full year (typical in petroleum refining) to periods of 12 hours on weekdays only (as in a carpet mill). Reliability is not necessarily related to maintenance. For example, whereas a petroleum refinery may be willing to completely rebuild a boiler once a year if the reliability is re-established, a carpet mill would expect virtually no maintenance costs for several years.

If reliability were affected by a maintenance problem (such as burner wear or cleanliness) it is a normal standard to have on-line replacement capability to restore performance. If the problem were continual, full-load capability may be expected even with maintenance being performed. It is normal to expect no maintenance problems with the steam generator in regard to tube life or material considerations. The typical industrial boiler customer prefers to have no steam-side maintenance, including circulation pumps or other items that could affect steam-generator reliability.

FLUIDIZED-BED COMBUSTION STEAM GENERATOR

The above analyses set the stage for a discussion of the application of fluidized-bed combustion (FBC) to industrial boilers.

More chemical energy can be converted in an FBC boiler than in conventional types. The outlook is that excess air values and combustible losses may be less than in other coal-burning techniques. In addition, if there is reduction of sulfur gases in the combustion products, a lower final temperature may be possible.

How the chemical energy is converted to useful thermal energy is somewhat different in an FBC boiler. Combustion takes place at a controlled temperature (now thought to be 1650°F for optimum pollution control). There is a great deal of discussion on the prediction of submerged heating surface. Some experimenters feel that the overall coefficient of 50 Btu/ft/°F, commonly assigned to the submerged heating surface, is too conservative. Suffice it to say that the mechanics of predicting bed temperature are not well defined. Radiation heat transfer capability from flue gases at the bed temperature as they traverse the combustion chamber signify that further temperature reduction of combustion products must be through convective heat transfer.

Using the previous outline, heating surface in the bed and in the walls of the fluid-bed chamber requires a system of steam-generating tube headers and steam drum, comparing to the water-cooled furnace in the convectional unit. Further utilization of the steam drum and separators, by adding parallel convective heat transfer surface, should be possible and is a factor in the temperature reduction of the combustion gases. It seems reasonable that an equivalent reduction to conventional designs can be made; so, a single heat trap would be sufficient.

Erie City has begun to develop FBC boiler designs to test certain features. A market survey of industrial boilers has confirmed a firm market position for steam generators having 250,000 lb/hr capacity, producing 750°F steam at 600 psig. To begin our analysis, we used these requirements as design basis. Also, we established that the physical configuration of the FBC steam generator would lend itself to shop assembly and to shipment by railroad.

We tried two different fluidized-bed designs. One utilized modular shop-assembled boiler units, with a fluid-bed chamber shipped separately from the boiler convection surface. The two would be joined together in the field.

In all arrangements, we proposed that combustion gases would flow in the freeboard area to one end of the chamber. They would be collected and passed through the convection zone. In one case, we considered sloped or vertical surface penetrating the grid and passing through the freeboard area. In another case, we considered only heating surface in the bed in the combustion chamber.

In still another trial, we set the goal as a totally shop-assembled unit. Where there are sloped tubes in the bed and the bed freeboard, we were able to duplicate conventional boiler performance if there was sufficient heating surface to have only one heat trap to produce the desired boiler efficiency. Table 2 compares performance values of the two-piece modular arrangement and of the single-piece unit. Although the advisability of the flow to one end may be questioned, it does have very real advantages. Heat transfer surfaces may be installed in the freeboard area and (most important) a particulate removal system may be installed to protect against excessive unburned carbon recirculation through both the convection pass and the heat trap. In accumulating all gases, convection pass heat transfer coefficients may be improved using conventional configurations of heating surface.

Figures 1 and 2 show two arrangements of FBC boiler components demonstrating the use of an intermediate particulate removing system.

So far, we have not mentioned transfer of heat to produce steam to elevate the useful energy that is transmitted to the process. It is fundamental that the superheater must be as carefully designed in FBC boiler applications as it is in conventional designs. Compared to conventional design, the fluid-bed system is largely an on-off device; consequently completely submerged superheater surface does not seem appropriate when low loads are considered. A combination of submerged in the bed and conventional surface appears to be better. Figure 3 shows one way to protect against excessive superheater submergence in the bed at low steaming rates: steam flow is biased toward the submerged surface to prevent overheating at low steaming capacity.

CONCLUSION

Reliability of the FBC industrial boiler appears to be the heart of the design problem. Although we have not designed the details of a fuel injection system or grid, we have set three goals:

1. The fuel injection system must have on-line replacement capability.
2. The grid system must have positive cooling to protect against temperature excursions approaching bed temperature.
3. The grid must be replaceable without major alterations to the steam generator.

We feel that the fuel feed systems and grid design are the principal hurdles to industrial steam generator customer acceptance of an FBC boiler design. We feel that the other objections can be overcome by a design that sacrifices shop assembly.

At this point, we are not at all confident that the FBC atmospheric steam generator will win acceptance over the present alternatives without full utilization of the potential for pollution control. With pollution control, however, lower cost and more readily available fuel supplies can be used. It is in this area, that air-fluidized-bed combustion techniques will have worthwhile economic compensations.

Table 2. COMPARISON OF DESIGN VALUES FOR TWO FLUIDIZED-BED COMBUSTION BOILER DESIGNS

Characteristic	Modular design (no freeboard heating surface)	Single unit (with sloped tubes in bed and freeboard heating surface)
Fuel	Coal	Coal
Steam flow, lb/hr	250,000	250,000
Steam pressure, psig	600	600
Steam temperature, °F	750	750
Boiler efficiency, %	87.7	87.7
Heat input, M Btu/hr	337	337
Heat absorption M Btu/hr		
Combustion chamber & superheater	195.6	244.0
Convection pass	68.5	20.1
Economizer	24.9	24.9
Total heat absorption	289.0	289.0
Gas temperatures, °F		
Leaving combustion chamber	1530	925
Leaving convection pass	672	672
Leaving economizer	350	350
Combustion air flow, lb/hr	275,000	275,000
Excess air, % of theoretical	10.0	10.0
Flue gas flow, lb/hr	298,000	298,000
Fluid bed press. drop, in. wg	40.0	40.0
Freeboard press. drop	.0	4.6
Convection press. drop	6.2	7.1
Total press. drop, in wg.	46.2	51.7
Gas velocity, ft/sec		
Through furnace	45	67
Entering convection pass	100	90
Superheater press. drop, psi	50	50
Heat transfer surface, ft^2		
Submerged in bed	3,200	3,200
Surface above bed	1,100	4,970
Convection pass	7,710	3,510
Superheater	860	970[a]
Total heat transfer surface, ft^2	12,870	12,650
Approximate height/length/width, ft	16/40/17	16/40/13

[a] Does not include 620 ft^2 in the bed.

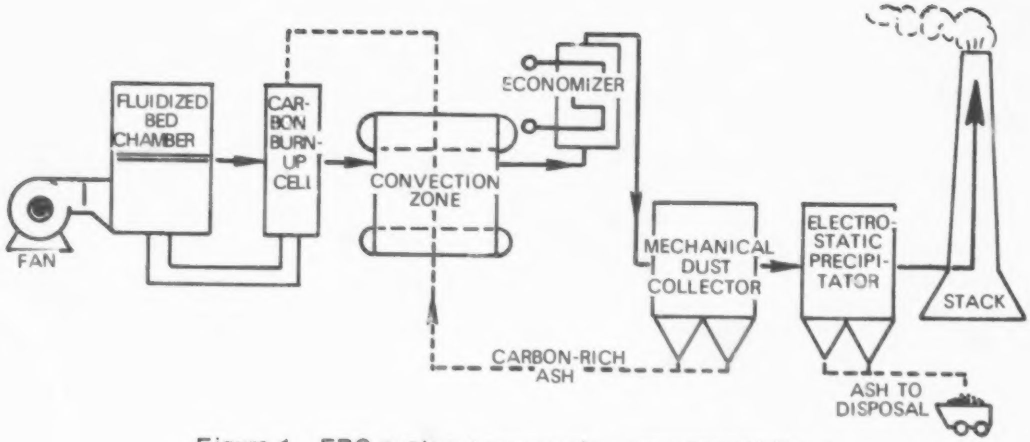
FLUIDIZED BED CHAMBER
CAR-BON BURN-UP CELL
CONVECTION ZONE
ECONOMIZER
MECHANICAL DUST COLLECTOR
ELECTRO-STATIC PRECIPI-TATOR
STACK
FAN
CARBON-RICH ASH
ASH TO DISPOSAL

Figure 1. FBC system components - arrangement No. 1.

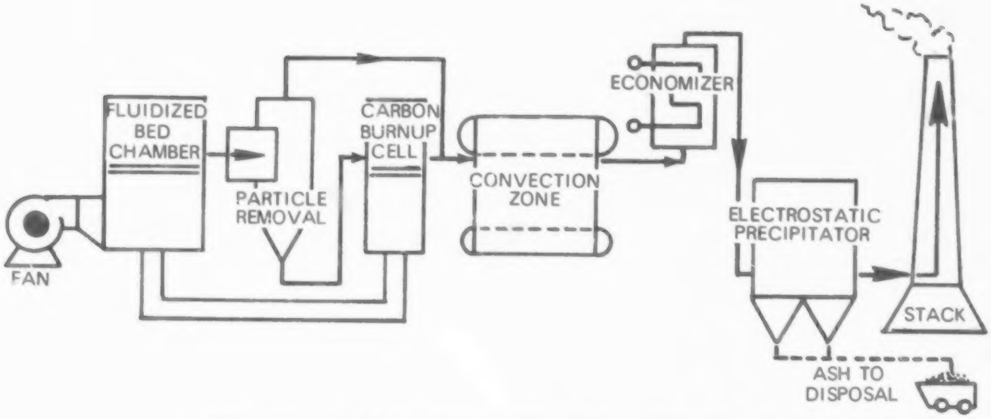
FLUIDIZED BED CHAMBER
PARTICLE REMOVAL
CARBON BURNUP CELL
CONVECTION ZONE
ECONOMIZER
ELECTROSTATIC PRECIPITATOR
STACK
FAN
ASH TO DISPOSAL

Figure 2. FBC system components - arrangement No. 2.

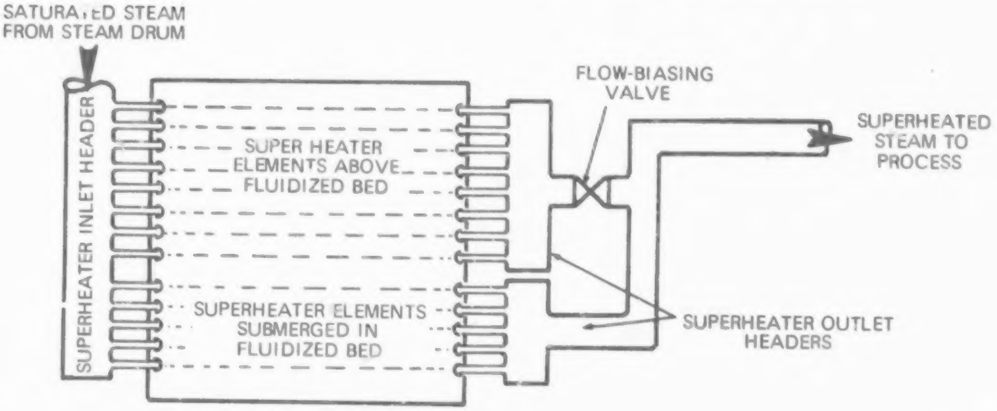
SATURATED STEAM FROM STEAM DRUM
SUPERHEATER INLET HEADER
SUPER HEATER ELEMENTS ABOVE FLUIDIZED BED
SUPERHEATER ELEMENTS SUBMERGED IN FLUIDIZED BED
FLOW-BIASING VALVE
SUPERHEATED STEAM TO PROCESS
SUPERHEATER OUTLET HEADERS

Figure 3. Superheater arrangement.

4. DESIGN FEATURES OF PRESSURIZED FLUIDIZED-BED BOILERS

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INTRODUCTION

As part of its effort on a NAPCA-funded study* to evaluate the fluidized-bed combustion process, Foster Wheeler is studying conceptual designs of fluidized-bed boilers. Although atmospheric-pressure fluidized-bed boilers for a 600-MW plant are also to be studied, work thus far has concentrated on pressurized boilers for a 300-MW plant. These pressurized 300-MW designs are the subject of this presentation.

ADVANTAGES OF PRESSURIZATION

Application of fluidized-bed combustion to a utility steam generator requires selection of a cycle and conceptual design which will make best use of the improved heat transfer offered by fluidization. Considering the relatively low average temperature at which combustion takes place in the bed, the combined steam-turbine/gas-turbine power plant with a pressurized boiler (see Figure 1) must be considered an immediate candidate. The higher heat transfer coefficients and high gas densities of pressurized operation should offer an excellent opportunity to reduce the first cost of the steam generator.

Pressurizing a conventionally fired boiler in this manner reduces the amount of heating surface required, which reduces its size and weight. Ultimately these reductions are reflected in a reduction in first cost as compared to a conventional non-pressurized boiler designed for the same steam conditions and net cycle output. The reduction is achieved as a result of an increase in emissivity of the non-luminous gases, higher gas densities and available pressure drop, higher permissible gas mass flows and convection heat transfer coefficients, and the reduced boiler duty requirement which results from splitting power generation between the gas turbine and steam turbine. Fluidized-bed combustion should offer further improvements in first cost by making use of high heat transfer coefficients throughout virtually the entire exchange of energy from the combustion gas to the steam, which is true when gas-turbine inlet temperature closely approaches the fluid-bed temperature.

CYCLE SELECTION

Cycle selection is ultimately dictated by requirements for gas side pressure and steam conditions.

Gas Side Pressure

Selection of the optimum gas side pressure is, of course, determined by studying the effect of pressure on overall plant economy. An examination by Westinghouse of the influence of pressure on plant heat rate and output indicates that there is little advantage in operating the gas turbine at boiler inlet pressures above 10 atmospheres. For a given boiler capacity, the cost of the pressure shell is most strongly related to gas pressure. Surface requirements are a function of steam cycle conditions and, except for some secondary effects, remain unchanged by gas side pressure. Effects of pressure on cost of auxiliaries such as coal handling equipment cannot be ignored. However, since they are beyond the scope of this presentation they will temporarily be overlooked.

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* Westinghouse is prime contractor on this program.

The cost of the shell is affected by pressure level, gas velocity, capacity, and diameter. The latter two variables are related in that the designer can resort to modular design at an appropriate plant capacity to minimize the effects of a further increase in shell size and to make use of shop fabrication techniques rather than field erection. Below a 12-1/2 ft diameter, a substantial reduction in cost can be realized by using shop fabrication. Modular construction, however, is not without bounds. When more than four or five vessels are considered, the effective reduction in vessel size diminishes rapidly, as shown in Figure 2. Gas velocity is more or less fixed by the combustion process. Any effect it may have on shell size is secondary compared to the other variables. As pressure level increases, shell size decreases rapidly due to the increase in gas density. Above 10 atmospheres, this effect on shell size diminishes, as shown in Figure 3. This diminished effect is compounded by the increase in shell thickness required to withstand the higher pressure. Considering that the incremental cost of auxiliary equipment, such as the coal handling system, also increases with an increase in pressure, 10 atmospheres appears to be an optimum pressure level.

Steam Conditions

The economics can be enhanced by applying the pressurized boiler to either the supercritical or subcritical pressure steam cycle and using the once-through principle. Doing so eliminates the large and costly steam drum required by a natural circulation boiler, as well as the risers and downcomers which would otherwise require numerous penetrations of the relatively small pressure containment shell. Once-through circuitry permits greater freedom in surface arrangement and location.

In factors influencing boiler cost, the subcritical cycle offers advantages (over the supercritical) of thinner wall tubes, lower tube metal temperatures, and some improvement in mean temperature between the gas and the steam.

Westinghouse thermodynamic and cost analyses of plants using subcritical and supercritical steam cycles combined with 10-atmosphere gas turbines also show that using supercritical steam conditions is not advantageous.

Based on the foregoing considerations, the design conditions of Table 1 were selected as a basis for the conceptual design of fluidized-bed boilers for a 300-MW plant. The approximate steam conditions in the boiler are shown superimposed on the temperature-enthalpy diagram in Figure 4.

CONCEPTUAL DESIGNS

Basic Criteria

Conceptual arrangements of the pressurized fluid-bed boilers are based on several fundamental criteria.

1. Where possible, modular construction is used to take the greatest possible advantage of shop fabrication techniques and (thereby) of lower cost.
2. A minimum of four completely self-contained modules per boiler facilitate the highest possible turndown ratio in the shortest permissible time.

3. The fluid beds are further divided by heating functions (i.e., pre-evaporative heating, evaporation, superheating, and reheating) to simplify startup procedures, reduce startup time, and improve control of the various heating functions.
4. Pre-evaporative heating and evaporation may be combined in a single bed, where vertical tubes are being considered to minimize tube bending and manifolding and to avoid possible tube overheating due to stratified flow of steam and water. (Under these circumstances a minimum of 12 beds are required of two different sizes.)

Pressurized operation offers the advantage of higher permissible pressure drops through the bed without appreciably affecting plant economy. This means it is possible to go to higher gas velocity and much deeper beds. For example, at 1 atmosphere and gas velocities of 4 ft per second, the bed may be about 2 or 2-1/2 ft deep. A comparable bed at 10 atmospheres and 15 ft per second could be 30 ft deep. Going to higher pressures changes the ratio of height to plant area considerably.

Going to high gas velocities runs the risk of erosion of tube surface, elutriation of large quantities of fly ash and carbon, and increasing the carbon loss. Instead, conceptual designs are based on stacked beds with gas velocities of about 4 ft per second; the volume occupied by the bed remains the same as if gas velocities of 15 ft per second were selected. This same volume is accomplished by splitting the air flow and putting four beds in parallel on the air/gas side rather than passing all the air through a single bed.

Preliminary Layouts

Figures 5 and 6 show two possible arrangements of the boiler. In both these arrangements, air enters the shell at the bottom of the vertical boiler and flows upward through the space between the shell and a finned tube water wall which encloses the boiler proper. The required portion of the air is drawn off at each bed level, and passed into a plenum chamber housing the tube headers, and through a grid plate into the bed. Gases leaving the fill space above each bed pass through an opening in the inner water-cooled wall into a central channel and thence downward out of the vessel. The gases are returned to the gas turbine in an air-cooled double-lined pipe in which the air from the compressor is the coolant.

This arrangement not only provides air-cooling of pressure parts, headers, and coal

Table 1. DESIGN CONDITIONS FOR COAL-FIRED, PRESSURIZED, FLUIDIZED-BED BOILER FOR 300-MW PLANT

Design Characteristic	Criteria
Primary steam flow, lb/hr	1,727,000
Superheater outlet pressure, psig	2500
Superheater outlet temperature, °F	1000
Reheat steam flow, lb/hr	1,644,000
Reheat inlet temperature, °F	650
Reheat outlet temperature, °F	1000
Reheat inlet pressure, psig	600
Reheat outlet pressure, psig	580
Feedwater temperature, °F	578
Fluidized-bed pressure, atm	10
Air flow to boiler, lb/hr	2,340,000
Air temperature to bed, °F	575
Fluid-bed temperature, °F	1700
Gas temperature leaving boiler, °F	1600
Bed-to-tube heat transfer coefficient, Btu/hr/ft^2/°F	50

conduits, but also ensures that in-leakage of air (rather than out-leakage of hot gas) will occur in the event of any breach in the finned tube enclosure wall. It also reduces the total external piping required by carrying the hot gases within the pressure vessel a part of the way to the gas turbine.

Annular beds are used in the Figure 5 arrangement, which has the advantage of making maximum use of the space available within the shell. It also separates zones of high differential pressure on the air/gas side with cylindrical surfaces which minimizes bracing and reinforcement.

Coal is introduced tangentially at a number of points on the outer periphery of the beds. Three injection points per bed should give adequate distribution to permit blowing the coal in through tubes located between the water tubes of the enclosure wall.

Tube bundles in the superheater and reheater sections consist of vertical, multi-start, helical coils that fill the annular beds. The vertical evaporating tubes are arranged radially in the annular bed and are connected to ring-type headers. The enclosure wall tubes serve as part of the pre-evaporator surface; the balance is within the evaporator bed.

Helical coils are advantageous for this application because of the relatively small number of tubes leading into and out of the bed. The same is true of flat spiral coils, which might also be employed. However, coils have the disadvantage of high fabrication cost. Also, it is impossible to replace individual tubes within a coil bundle; the only recourse is to plug the ends of leaking tubes.

The arrangement of Figure 6 uses rectangular beds and the more conventional horizontal return-bend tube elements in the superheater and reheater. This arrangement provides for easier maintenance but has the disadvantage of large flat surfaces which have to be reinforced to withstand gas differential pressures. Space utilization within the shell is not as efficient as with annular beds.

Cyclones and dip legs (not shown in Figures 5 and 6) are in the free space above beds to collect elutriated material and return it to the beds.

Note that the just-discussed concepts are preliminary. They are two of several bed-and-surface arrangements that will be critically evaluated prior to developing a single detailed design. Other concepts include a single deep bed in each vessel, and horizontal vessels containing relatively shallow beds. Much more detailed study of the manufacturing, assembly, and maintenance problems involved in these arrangements is required.

Surface Requirements

Heating surface area and bed volume requirements of the boiler have been calculated by assuming that 2-in. O.D. tubes are used throughout and that the tubes in the bed are placed on an average spacing equivalent to a 4-in. square pitch. Although 2-in. O.D. tubes throughout may not be optimum, they are a reasonable choice, considering problems of support, possible vibration, and the number of circuits and tube-to-heater connections. It has also been conservatively assumed that the bed-to-tube heat transfer coefficient is constant throughout the bed at 50 Btu/hr/ft^2/°F, regardless of tube orientation. Table 2 gives the results of calculations for the annular bed arrangement shown in Figure 5. For the rectangular bed arrangement of Figure 6, the amount of pre-evaporator surface in the bed would be reduced because of the greater amount of enclosure wall surface.

Using smaller than 2-in O.D. tubes of the same pattern and pitch-to-diameter ratio reduces bed volume considerably. For example, for 1-1/2 in. tubes on 3-in. centers and 1-in. tubes on 2-in. centers, the bed volume will be about three-quarters and one-third as much, respectively, as for 2-in. tubes on 4-in. centers.

COMPARISON WITH CONVENTIONAL BOILER

Table 3 compares a pressurized fluidized-bed boiler to a conventional boiler for the same steam conditions and plant output. The maximum mean tube wall temperatures are nearly the same in both units. The cost of material in the superheater and reheater of the fluidized-bed boiler is only about one-fifth of that in the conventional boiler. Surprisingly, no austenitic steel is required in the reheater. All this, however, is based on the relatively conservative bed-to-tube heat transfer coefficient of 50 Btu/hr/ft^2/°F. Any increase in this coefficient changes the material picture considerably.

Table 2. CALCULATED SURFACES AND BED VOLUMES—PRESSURIZED, FLUIDIZED-BED BOILER FOR 300-MW PLANT[a]

Component	Heating surface, ft^2	Bed volume, ft^3	Overall H.T. Coeff, Btu/hr/ft^2/°F
[b]Pre-evaporator			
[c]Enclosure walls	10,500	–	20[d]
Immersed in bed	3,300	700	47
Evaporator	9,780	2,070	47
Superheater	15,820	3,360	43
Reheater	8,350	1,770	39

[a] All values are approximate.
[b] Enclosure wall length based on assumed 15-ft free height above each separate bed.
[c] Portions of the enclosure walls are immersed in the beds.
[d] Weighted average.

Table 3. COMPARISON OF CONVENTIONALLY FIRED AND PRESSURIZED FLUIDIZED-BED STEAM GENERATORS FOR 320-MW PLANT OUTPUT

Characteristic	Conventional	Fluid-bed
Primary steam flow, lb/hr	2,189,000	1,727,000
Superheater outlet pressure, psig	2,500	2,500
Superheater outlet temperature, °F	1,000	1,000
Reheat steam flow, lb/hr	1,936,000	1,644,000
Reheat inlet temperature, °F	650	650
Reheat outlet temperature, °F	1,000	1,000
Reheat inlet pressure, psig	600	600
Reheat outlet pressure, psig	580	580
Feedwater entering steam generator, °F	486	578
Superheater		
Surface area, ft^2	61,660	15,820
Max. mean tube wall temperature, °F	1,095	1,078
Material weights, lb		
Carbon steel	108,000	36,000
C–1/2% Mo		24,000
1/2% Cr –1/2% Mo	62,000	17,000
1% Cr –1/2% Mo		19,000
1-1/4% Cr –1/2% Mo	73,000	22,000
2-1/4% Cr – 1% Mo	430,000	22,000
18 Cr – 8 Ni	20,000	7,000
Total material weight, lb	693,000	147,000
Relative material cost, %	100	19
Reheater		
Surface area, ft^2	67,800	8,350
Max. mean tube wall temperature, °F	1,115	1,136
Material weights, lb		
Carbon steel	140,000	
1/2% Cr – 1/2% Mo	133,000	
1-1/4% Cr – 1/2% Mo	59,000	24,000
2-1/4% Cr – 1% Mo	64,000	14,000
9% Cr – 1% Mo		21,000
Total material weight, lb	396,000	59,000
Relative material cost, %	100	18

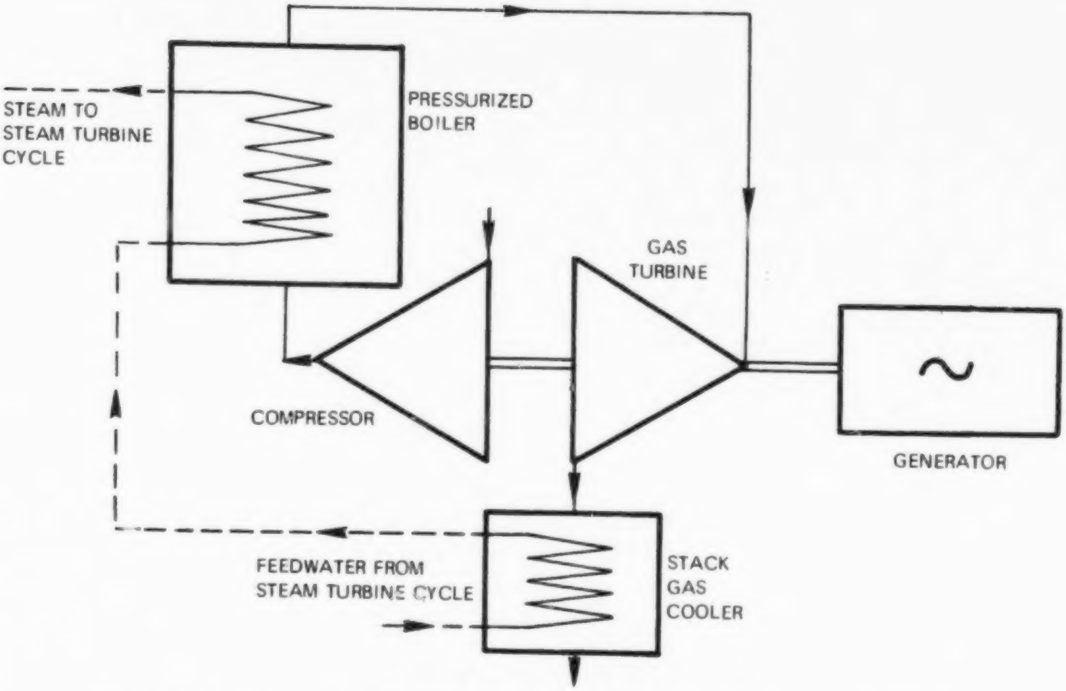
STEAM TO
STEAM TURBINE
CYCLE
PRESSURIZED
BOILER
GAS
TURBINE
COMPRESSOR
GENERATOR
FEEDWATER FROM
STEAM TURBINE CYCLE
STACK
GAS
COOLER

Figure 1. Combined steam-turbine/gas-turbine cycle with pressurized boiler.

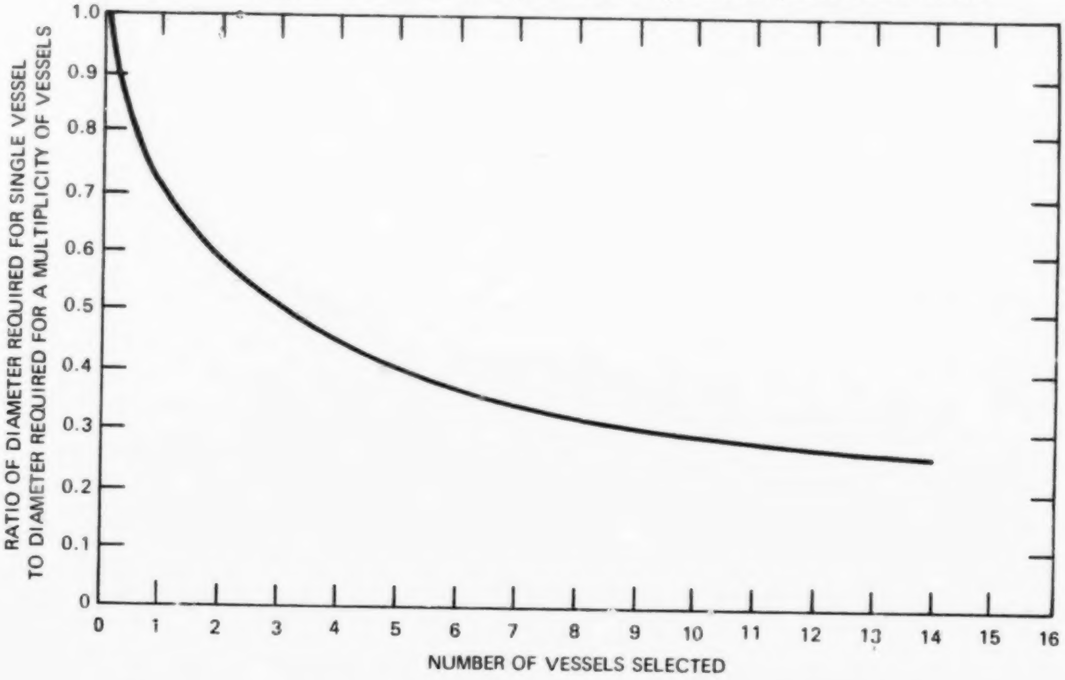

Figure 2. Effect of the number of vessels selected on the vessel size.

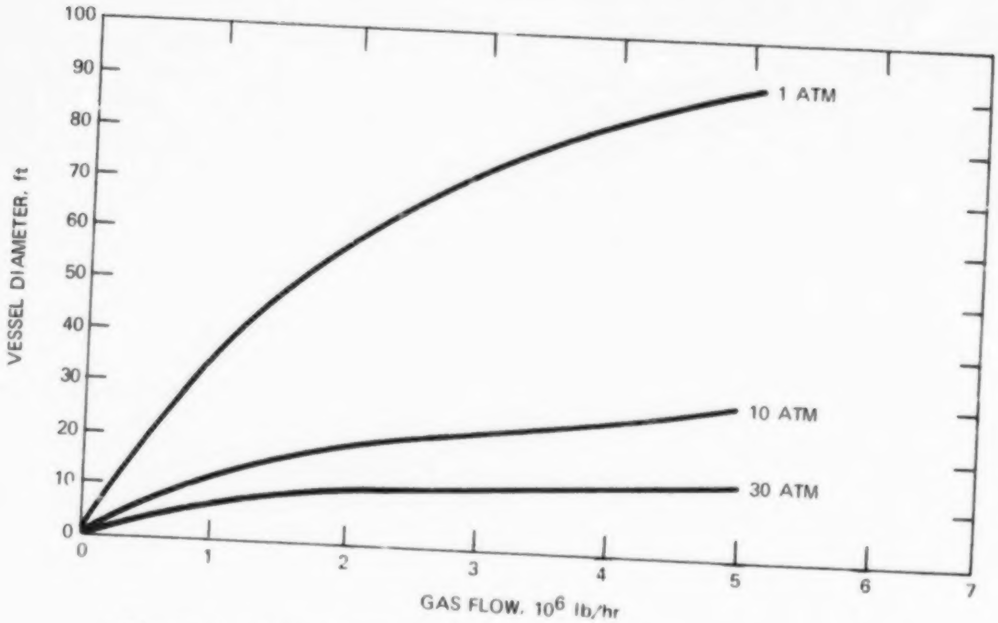

Figure 3. Effect of gas flow and pressure on vessel diameter.

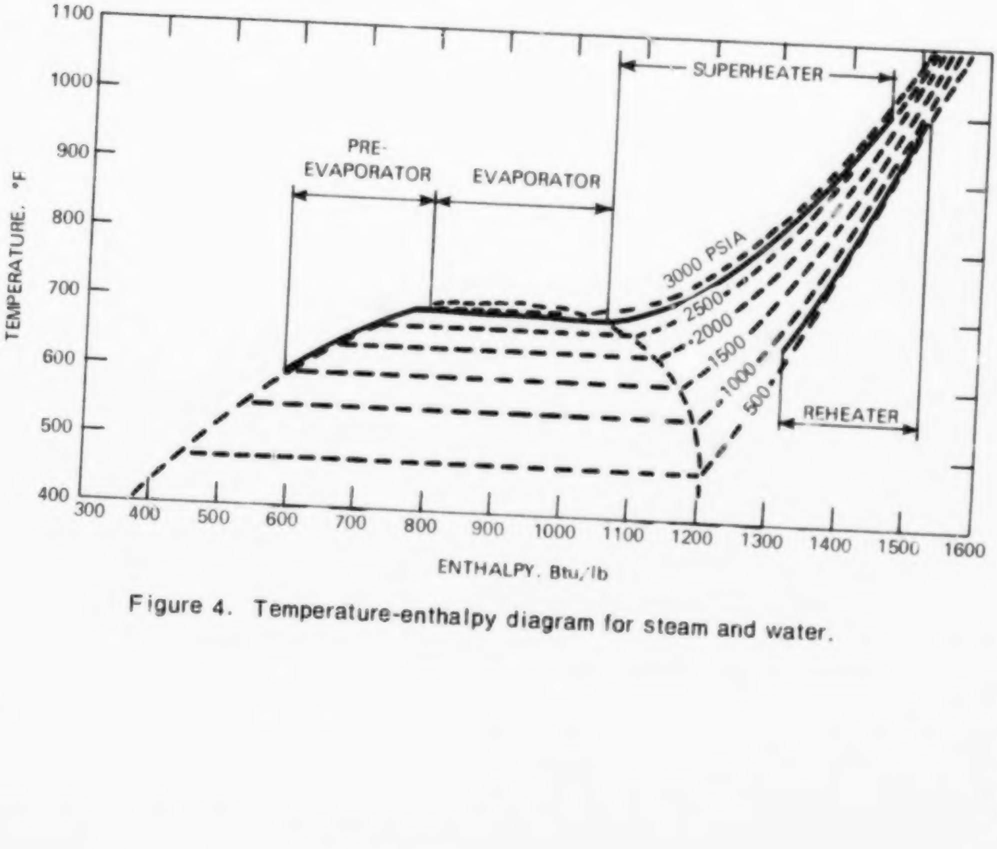

Figure 4. Temperature-enthalpy diagram for steam and water.

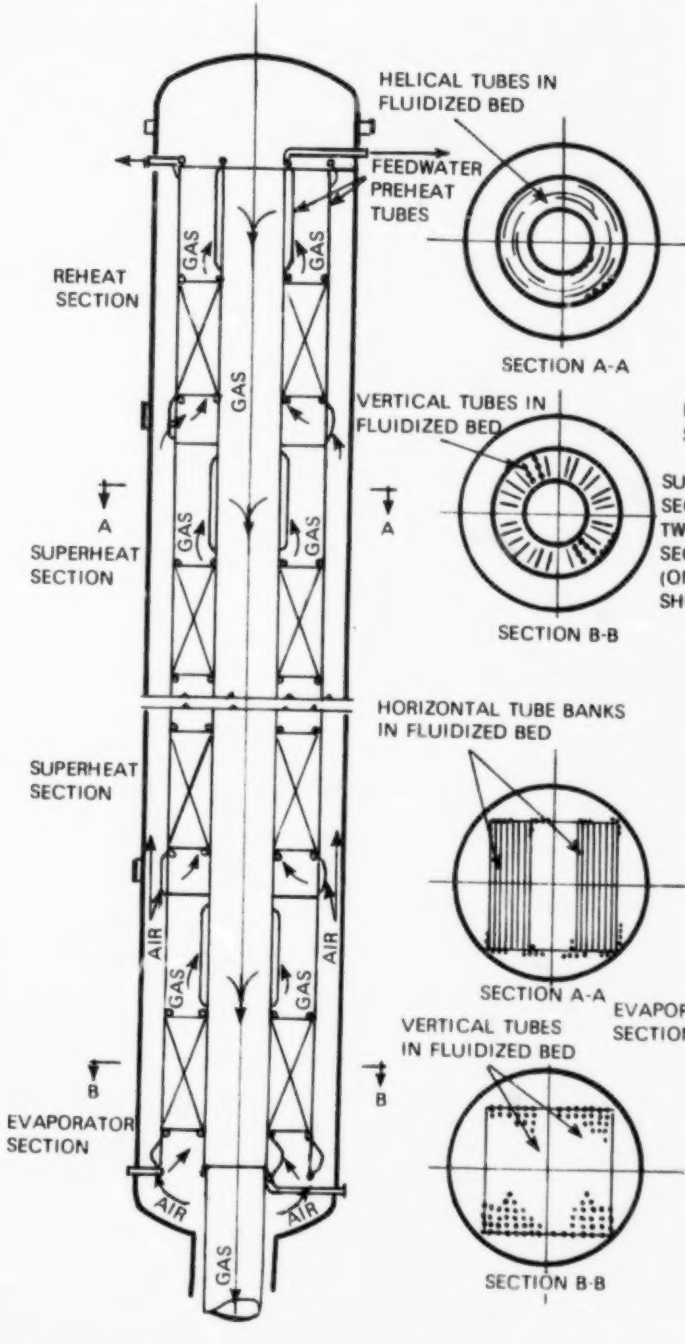
HELICAL TUBES IN FLUIDIZED BED
FEEDWATER PREHEAT TUBES
GAS
REHEAT SECTION
SECTION A-A
VERTICAL TUBES IN FLUIDIZED BED
A
SUPERHEAT SECTION
SECTION B-B
SUPERHEAT SECTION
AIR
B
EVAPORATOR SECTION

Figure 5. Pressurized fluidized-bed boiler -- annular beds.

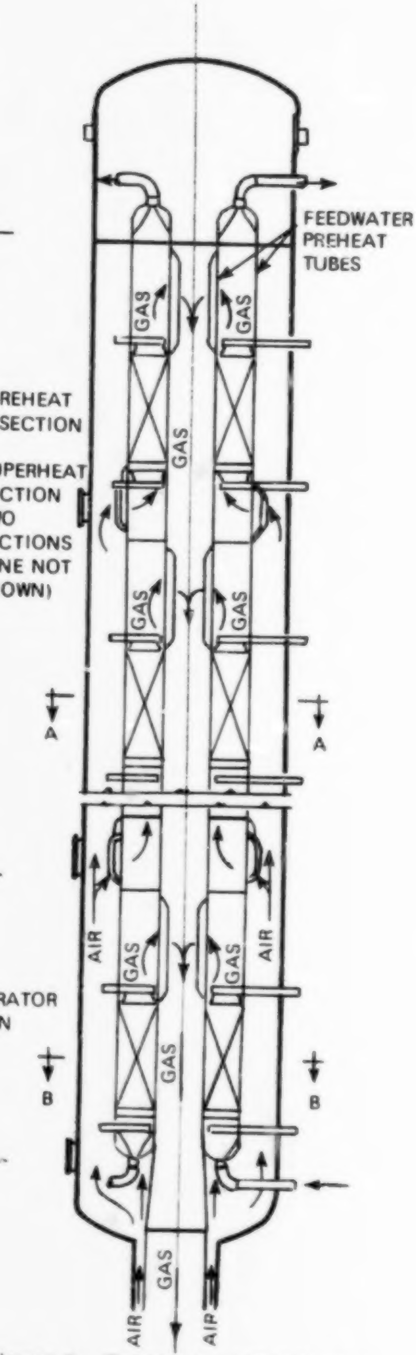
FEEDWATER PREHEAT TUBES
GAS
REHEAT SECTION
SUPERHEAT SECTION TWO SECTIONS (ONE NOT SHOWN)
HORIZONTAL TUBE BANKS IN FLUIDIZED BED
SECTION A-A
A
EVAPORATOR SECTION
VERTICAL TUBES IN FLUIDIZED BED
SECTION B-B
B
AIR

Figure 6. Pressurized fluidized-bed boiler -- rectangular beds.

5. SOME ECONOMIC ASPECTS OF HIGH-TEMPERATURE STEAM CYCLES

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INTRODUCTION

One of the most important aspects of fluidised-bed combustion is the very low corrosion rates which occur when metals are immersed in the solids; hitherto, steam cycles have been limited to a temperature of 1050°F because of fouling and corrosion of superheater tubes. Thus the heat rate of steam plant, which showed a downward trend until the mid 1950s, has now flattened out at about 2.5 kW heat/kW electricity. As fluidised-bed combustion may allow us to continue the downward trend of the heat rate curve, we have undertaken a brief examination of the cost of achieving better steaming conditions.

FOULING AND CORROSION

Dainton and Elliott[1] immersed a typical stainless steel probe for 120 hours in a fluidised bed burning Thoresby coal at a temperature of 1300°F. This coal, notoriously viscous, has a high chlorine/sulphur content and would cause a normal power station to shut down within a day because of savage fouling. The probe was cooled internally by air so that the exterior in contact with the ash varied in temperature from 900 to 1200°F. The metal surface which was at a temperature above 1100°F was clear of any deposit or corrosion; at temperatures below 1100°F, fouling became progressively worse as the temperature decreased. The deposit, mainly sodium chloride, was accompanied by a small amount of surface pitting. By contrast, the fouling of superheater tubes in a test rig at B.C.U.R.A., simulating pulverised fuel firing and using the same coal, gave massive deposits which almost bridged across the tube bundle in a 24-hour experiment.

As the absence of fouling and corrosion is one of the cornerstones of our present proposals, it is worth looking at some of the fundamentals involved. Figure 1 shows the variation with temperature of the vapour pressure of some of the alkali metal salts present in most coal ashes. It will be seen that the vapour pressure levels at normal flame temperatures of about 2500°F are several orders of magnitude higher than those in a fluidised bed operating at 1500°F. As fouling is a process of deposition from the vapour phase, it is reasonable to predict much lower fouling rates at the lower temperatures. Figure 1 also shows that the vapour pressure of sodium chloride at 1300°F is significantly higher than that of most of the other compounds and would therefore be the one most likely to be deposited; this agrees with experimental results. It is interesting to note that the vapour pressures of the sulphates, which are a common cause of fouling in normal boilers, is negligible under fluidised-bed conditions.

The absence of fouling on the metal surfaces which are above 1100°F can be likened to the phenomenon of deposition of dew from moist air. Unless there is a sufficient temperature differential, little deposition occurs; if there is any, it could well be removed by the mildly abrasive action of the solids. Only when there is a substantial temperature difference does the deposit adhere. It should be pointed out that the experiments were conducted with very fine coal at low fluidising velocities; we could expect less deposits when using larger coal, unless the nature of the fluidising conditions gives rise to particle temperatures well in excess of bed temperature. When similar trials were undertaken with coals which are considered to be bad (but tolerable) to use in normal systems, no fouling whatsoever occurred at any metal surface temperature.

These preliminary encouraging results have been supported by experience in the larger combustors now running; although we need more evidence from much longer duration experiments, it is useful to enquire if the potential to go to higher temperatures would be worthwhile economically.

STEAM CYCLES

Four steam cycles have been investigated: the first two were based on the design conditions of an existing supercritical p.f.-fired power station; the other two were cycles considered by Downs.[2] Table 1 gives working conditions and the efficiency of these cycles, together with those of a standard 500-MW set which has been taken as datum. It is not suggested that these cycles would be adopted in an actual design; however, they represent (at the lower end) plant which could be achieved without departing from current steam turbine technology and (at the upper end) plant which could possibly be developed in the next decade.

FLUIDISED-BED CONDITIONS

Tube cost calculations are based on the fluidised-bed operating conditions shown in Table 2. These conditions were deliberately chosen on the pessimistic side; e.g., we have evidence that heat transfer coefficients in excess of 70 Btu/ft²/hr/°F can be achieved, and it is likely that operating the bed above 1500°F would produce a more economical tube bundle.

In each case it is assumed that all the superheater and reheater tubes are in the fluidised bed to take full advantage of reduced heat transfer surface. Cooling of the gases after the fluidised bed is by normal convective surfaces. The assumed fluidising velocity is rather low (2 ft/sec) to correspond to conditions in a pressurised system; however, the same argument would hold for higher velocities.

TUBING DESIGN

A wide range of high-temperature alloys have been developed for use in gas turbines which may prove suitable for boiler application; the final choice would most likely be made because of weldability and resistance to internal waterside corrosion, rather than to optimise the cost of the tubing. Nevertheless the cost of tubing to perform a given duty can be estimated as follows:

1. For each metal considered, calculate values of tube thickness for metal temperatures in the range 572-1472°F (300-800°C), using the formula:

$$t = \frac{d_i P}{2f - P} + \frac{40 (2f + P)}{P (2f - P)}$$

where,

t = wall thickness (in.) but never less than 0.092 in.
d_i = internal diameter (in.)
P = working pressure (psig)
f = working stress of metal at the appropriate temperature in psi.

Table 1. ASSUMED STEAM CYCLE CONDITIONS

Cycle No.	Point of comparison / Cycle[a]		Generated power, MW	Steam pressure, psi	Boiler working fluid conditions				Steam flow rate, Mlb/h	Steam cycle efficiency, %	Assumed sent out peak efficiency, %	Reduction in fuel consumption, %
					Temperature		Enthalpy					
					Inlet, °F	Outlet, °F	Inlet, Btu/lb	Outlet, Btu/lb				
Datum	Fluidised-bed 500-MW cycle	(a)	500	2300	493	1050	476	1488	3.4	42.8	39.5	0
		(b)		585	689	1050	1344	1545	2.66			
1	Drakelow C	(a)	375	3500	505	1100	498	1495	2.405	45.9	42.4	7.35
		(b)		735	683	1050	1324	1540	1.794			
2	Logical single-reheat	(a)	464	3500	500	1300	488	1629	2.405	47.9	44.2	11.9
		(b)		770	700	1250	1341	1651	1.804			
3	Optimum-pressure single-reheat	(a)	460	6500	500	1300	489	1572	2.405	48.6	44.8	13.4
		(b)		1430	700	1250	1292	1638	1.804			
4	Optimum-pressure double-reheat	(a)	528	6500	500	1300	489	1572	2.405	49.4	45.6	15.4
		(b)		1430	700	1250	1292	1638	1.804			
		(c)		286	700	1250	1369	1661	1.353			

[a] Each cycle shows separate conditions for: (a) main steam; (b) reheater No. 1; and (c) reheater No. 2.

The second term in the formula is a corrosion allowance; vary it according to experience.

2. Using t, and knowing the fluidised-bed temperature, θ_b, calculate the overall bed-to-steam heat transfer coefficient, h, assuming:
   (a) a bed-to-tube heat transfer coefficient of 70 $\text{Btu/ft}^2\text{/h/}^\circ\text{F}$,
   (b) a tube-to-steam heat transfer coefficient of 500 $\text{Btu/ft}^2\text{/h/}^\circ\text{F}$, and
   (c) values of thermal conductivity for the material under consideration.
3. Knowing θ_b and h, calculate the working fluid temperature θ_f.
4. Knowing θ_b, t, h, θ_f, and the metal density, determine the relative weight of material to transfer one unit of heat at the appropriate temperature. If the cost of the metal and its cost of fabrication is estimated, the cost to transfer a unit of heat can be calculated. This allows the cheapest metal to be chosen for any given steam condition. Normally it would not be desirable to use more than four different materials in one boiler in order to minimise the number of dissimilar welds. Figure 2 shows how the relative costs to transfer a unit of heat would appear using 2-1/4 percent chrome steel, Austenitic 316, and Nimonic PE16 with costs per ton of £750, £3000, and £7500, respectively, for steam pressures of 3500 psig, and using 1-in. I.D. tubes.
5. Finally, calculate the individual costs of metal for the different sections of the boiler tubing for the various cycles. Table 3 gives the results, using the above materials and mild steel.

Table 2. ASSUMED FLUIDISED-BED OPERATING CONDITIONS

Characteristic	Condition
Combustion bed temperature, °F	1472
Gas temperature at inlet to air heater, °F	662
Waste gas temperature at input to stack, °F	248
Fluidising velocity, ft/sec	2
Excess air, %	20
Base plate pressure drop, in. wg	5
Bed depth, ft	2
Density of fluidised bed, lb/ft^3	50
Bed-to-tube heat transfer coefficient, $\text{Btu/ft}^2\text{/hr/}^\circ\text{F}$	70
Tube-to-working-fluid heat transfer coefficient, $\text{Btu/ft}^2\text{/hr/}^\circ\text{F}$	500
Tube internal diameter (I.D.), in.	1

For comparison, the cost of the tubes in the datum standard 500-MW, 2300 psi x 1050°F x 1050°F set, using the same fluidized-bed conditions would be £ 0.6/kW. (We have chosen to compare the advanced cycle conditions with those of a fluidised-bed boiler; we believe the latter will prove more economic than normal pulverised-fuel systems.)

In addition to the increased boiler tubing cost when going to the higher steaming conditions, there is also an increase in the cost of the economiser tubing, due to the higher operating pressure. This increase has been allowed for in the overall comparison shown later.

FUEL COST SAVINGS

The present-day worth of the savings in fuel has been calculated assuming an equivalent life time load factor of 50 percent, a

Table 3. CAPITAL COSTS[a] OF 1-IN. BOILER TUBES

Tubing	Cycle 1	Cycle 2	Cycle 3	Cycle 4
Main and superheater				
Nimonic PE16	0.24	0.74	2.28	1.92
Austenitic 316	0.20	0.17	–	–
2-1/4% chrome steel	0.18	0.11	–	–
Reheater				
Nimonic PE16	–	0.17	0.18	0.26[b]
Austenitic 316	0.02	0.06	0.08	0.11[b]
2-1/4% chrome steel	0.03	0.03	0.03	0.05[b]
Mild steel	0.02	0.01	0.01	0.01[b]
Total	0.69	1.29	2.58	2.35

[a] Costs given in £/kW.

[b] Figures include both reheaters.

station life of 30 years, and a return on capital of 7-1/2 percent for fuel costs of 3d/therm and 5d/therm. Table 4 shows these savings together with tubing and other costs.

OTHER COST SAVINGS

The importance of overall efficiency to the overall cost of a power station is not always appreciated. The cost of access roads, coal storage, conveyor and mixing systems, coal bunkers, mills, fans, gas cleaning plant, chimnies, foundations, services, cooling water supplies, condensers, and water purification plant, all costs come down pro-rata with the lowering of the heat rate. Since these items constitute more than half the total station cost, "other savings" are indeed most important. The converse (decrease of efficiency leads to increased capital cost) is also true, as has been found in numerous studies aimed at producing a cheap peak-load power station by cutting out fuel-saving systems; e.g., feed heaters and high-temperature superheaters.

DISCUSSION

Table 4 shows that the value of the fuel cost savings far outweighs the increased cost of the high-temperature boiler tubing and, together with the other savings, should allow more than enough margin to pay for the increased cost of the steam turbine. Note that, although we could realize a moderate increase in turbine inlet temperature, say to 1100°F, without altering turbine design, an increase to 1200°F would almost certainly require a change in construction of the steam turbine rotor. This would be a more difficult problem than the design of the turbine blading which could rely heavily on gas turbine practice. However, there would not appear to be any insuperable problems in ultimately producing 1300°F machines.

Although the estimates made in this preliminary investigation are not precise and until a detailed design study is made, we could not indicate the optimum conditions; there seems to be a good case for serious consideration of high-temperature systems. In addition to

Table 4. SAVINGS[a] IN CAPITAL AND CAPITALISED REVENUE RELATIVE TO DATUM CYCLE

Characteristic	Datum 500-MW unit	Savings			
		Cycle 1	Cycle 2	Cycle 3	Cycle 4
Boiler tubing costs	0.6	-0.09	-0.69	-1.98	-1.75
Economiser tubing costs	0.6	-0.1	-0.1	-0.5	-0.5
Other station costs[b]	23	+1.8	+2.7	+3.0	+3.6
Fuel costs					
3d/therm	72.9	+3.8	+5.9	+6.6	+7.5
5d/therm	110.1	+6.4	+9.9	+11.0	+12.5
Total savings					
3d/therm	–	+5.4	+7.8	+7.1	+8.8
5d/therm		+8.0	+11.8	+11.5	+13.8

[a] Savings given in £/kW.

[b] Most likely an underestimate; the full costs influenced by efficiency have not been completely examined.

design studies, we need further experimental evidence of both the internal and external corrosion resistance of high-temperature materials; accordingly, it is recommended that in the next round of pilot-scale plant, opportunity is taken to incorporate at least some tubes running under advanced conditions. Only if we can show evidence of good possibility of achieving high temperatures will turbine manufacturers be justified in undertaking the expensive development of advanced machines.

One interesting aspect of such development would be the result of an increased demand for high-temperature metals: would it create a shortage and a higher price, or lower material costs? Our calculations suggest that, even with the very advanced conditions, about 600 tons of Nimonic (or other equivalent high-temperature) material would be used in a 2000-MW station. Comparing this with the 20 tons of such alloys which are needed for the engines of a Concorde, it would appear that, although the demand would be large enough to justify quantity production, it is unlikely to produce a scarcity market.

CONCLUSIONS

1. The incremental cost of the tubing needed to achieve high-pressure high-temperature steam in a fluidised-bed boiler is relatively low: a 6500 psi x 1300°F x 1250°F x 1250°F boiler should not cost as much as a conventional pulverised-fuel-fired 2300 psi x 1050°F x 1050°F boiler.

2. The fuel cost savings which would accrue from these higher efficiency cycles are substantial; they would more than pay for the extra cost of the high-temperature steam turbine.

3. In addition to the reduced fuel costs, there should also be a saving in capital costs associated with the higher efficiency, because all plant items related to fuel throughput have their costs spread over a greater output.

4. Modest increases in steam temperature (within the existing capabilities of steam turbines) could be the prime objective of pilot-scale plant because they should present no extra development problem for the fluidised-bed boiler. Opportunity should also be taken to incorporate much higher temperature trial sections in such plant in order to assess the capabilities of such systems.

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ACKNOWLEDGEMENT

The authors wish to thank Mr. D. H. Stockwell of the Coal Research Establishment for carrying out detailed analyses and for general assistance in the preparation of this paper. They also wish to thank the National Coal Board for permission to publish the work. The ideas and conclusions expressed are those of the authors, not necessarily of the Board.

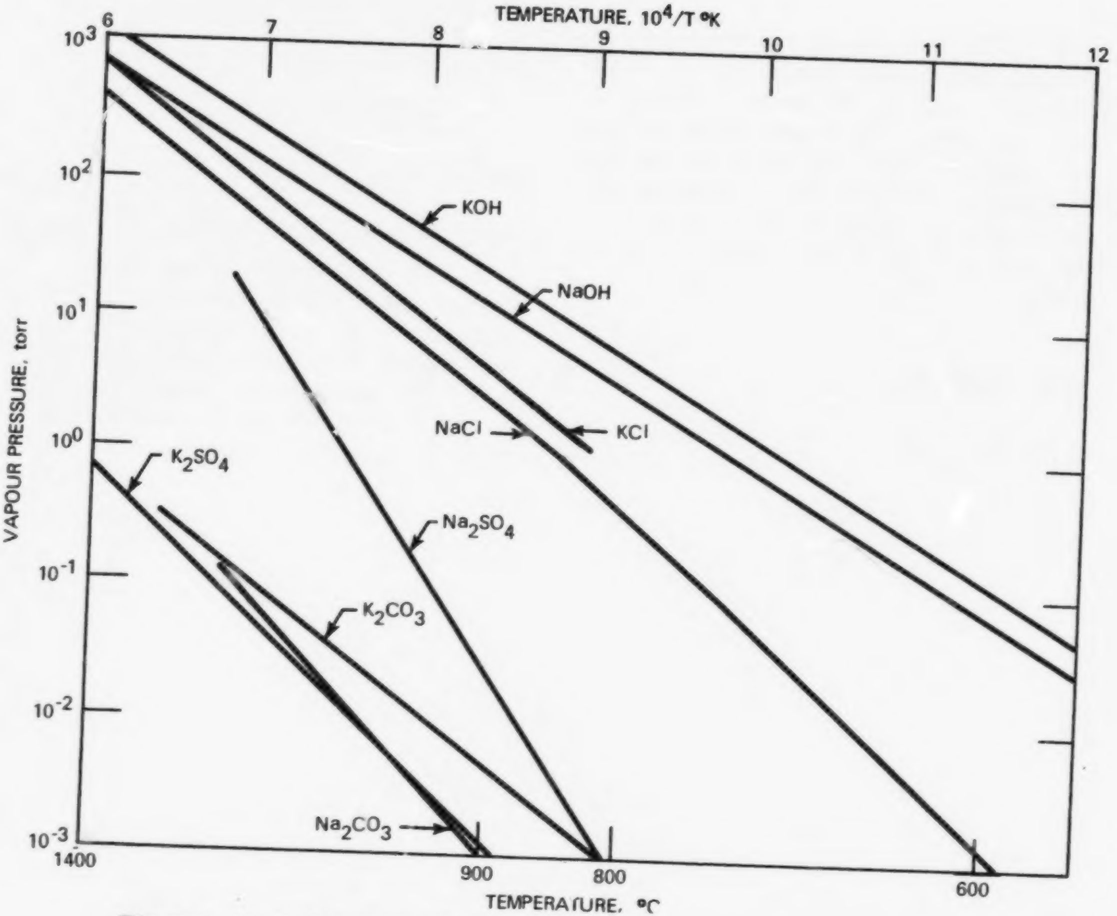

Figure 1. Variation (with temperature) of vapour pressure of alkali salts.

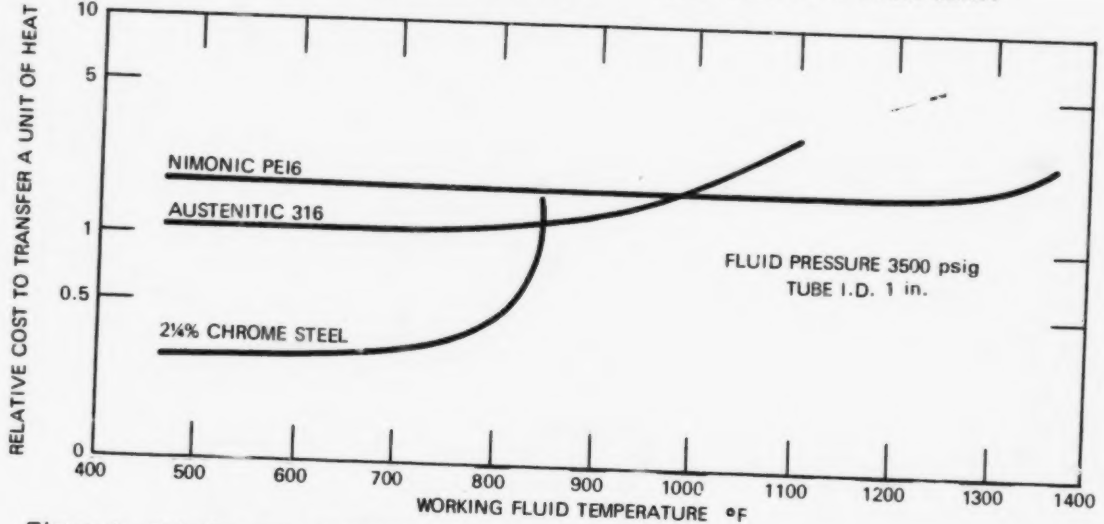

Figure 2. Relative cost of tubing for the high-pressure conditions of cycles No. 1 and 2.

6. COMBUSTION OF OIL OR GAS IN FLUIDIZED BEDS

L. REH

Lurgi Gesellschaft fuer Chemie und Huettenwesen GmbH

ABSTRACT

Over the last 10 years, Lurgi has built numerous plants for combustion of liquid refinery wastes with additional heating by refinery waste gas. Bad experiences during startup of the first plants led to a fundamental study of the combustion of liquid and gaseous fuels in a fluidized bed. The principal results of this work are discussed in this paper.

INTRODUCTION

About 10 years ago, the Lurgi Company had to solve a problem for several German refineries: burning liquid wastes from their waste water treatment plants. Because of the ever-changing net heating value of these viscous waste sludges, the fluidized-bed principle was chosen.[1,2,3]

COMBUSTION OF LIQUID REFINERY WASTES

The plants, built as compact units, had fluidized-bed furnaces of approximately 4.5 ID. The furnaces burned, for instance, 1400 lb/hr oil sludge with oil content of 12 percent, water content of 70 percent and inorganic matter content of 18 percent. The composition often changed rapidly, so that nearly all oil-contaminated water had to be "burned" and the heat requirement of combustion had to be supplied by injected gaseous fuel.

Figure 1 shows the principal flow scheme of such a plant. The incoming air was first preheated by the flue gases of the fluidized-bed furnace to 900°F and then split into primary and secondary air streams. The first was sent through the grate as fluidizing air to a sand bed into which the oil sludge was pumped; the second was blown at a very low angle of inclination over the fluidized-bed surface level. When necessary, gaseous fuel was injected over nozzles in the side wall of the fluidized-bed furnace at a distance not too high above the grate. The temperature of the fluidized bed was held in the 1500°F range; however, even with means for distribution of fuel into different fluidized bed sections, the temperature of the furnace off-gas ranged up to 1650°F under normal operating conditions, due to afterburning above the sand bed. The flue gases, completely freed from organic matter, passed through a tube-and-shell heat exchanger. The entrained ash was precipitated in a following cyclone before the gases left for final dust cleaning and disposal via the stack.

Figure 2 shows that, between net heating values of 2000 and 2500 Btu/lb of sludge, the optimal combustion temperature can be reached without any addition of gaseous fuel (which is necessary for lower net heating values) and without any cooling (needed for higher heating values).

When, for instance, an oil sludge with nearly no heating value was burned, heavy afterburning (caused by gaseous fuel) occurred in the upper part of the furnace. The bed temperature went down and the flue gas temperature up, sometimes leading to damage in the heat exchanger. The same occurred when light volatilizing fuels were burned.[4]

This problem led us to the study of the influence of the radial mixing of gases in a fluidized bed on combustion.

INFLUENCE OF RADIAL GAS MIXING ON COMBUSTION

In cooperation with the Technical University, Karlsruhe, Germany, first tests were made with cold models under similar fluidization conditions as in an actual fluidized-bed furnace. One of the cold models was the same size as a 32-in. diameter fluidized furnace which could be fired with gaseous fuels, diesel oil, or heavy oil.[5]

For the conditions existing in a fluidized bed with grate tuyeres with horizontally blowing openings, we studied, for instance, the injection length of the jet into a nearly two-dimensional fixed bed of sand of approximately 1/20-in. particle diameter. Even at high velocities (in the region of several hundred feet per second) the jet reached only the middle of the model at a width of approximately 1 ft; in the experiment, the influence of the jet affected the whole width of the sand bed, 2 ft above the injection point.

Following the model tests, a fluidized furnace was constructed, having a grate from seven horizontally blowing tuyeres into which gaseous fuel could be introduced either by the central tuyere in the furnace axis through the grate or by injecting nozzles by the side; the side nozzles could be used also for liquid fuels. Because premixed air/fuel gas mixtures, entering a fluidized bed by adequate grates to avoid back-ignition, give complete combustion, only those cases were studied in which mixing and combustion of fuel gases occur inside the bed.

Concentration and temperature profiles were measured at different heights of the fluidized-bed furnace. The samples were taken symmetrically to the injection points of fuel in two vertical planes rectangularly arranged to each other. By means of balance calculations, the mean content of unburned carbon in the fluidizing gas was found in every horizontal layer of the furnace.

Figure 3 is the temperature profile for a test, where town gas, injected through the central tuyere of the grate, was burned in a sand bed (1/20-in. mean grain diameter) with 50 percent excess air at a fluidizing velocity of approximately 5 ft/sec. The profile, typical for other measurements, shows clearly that the temperature in the bed adjacent to the wall (where, preferably, the air should be passing) is considerably lower than in the centre of the bed. Some combustion is still taking place above the bed level, regulated to 60 in. above the grate.

Combustion inside the bed is strongly dependent on the fluidization velocity, as shown in Figure 4, in which the content of unburned carbon for the aforementioned combustion conditions is plotted versus the axial coordinate of the sample point above the grate. At higher fluidization velocities, higher bed heights are needed to reach complete combustion because of a limited radial mixing rate, which also means longer mixing ways in the direction of gas flow. Especially at higher fluidization velocities, the content of unburned carbon leaving the fluidized bed at a bed level 60-70 in. above the grate is rather high; it reached values of 15-20 percent.

The concentration profiles, which will be published[5] later, indicate an excess of air in the bed adjacent to the wall; this excess is due to insufficient radial mixing, and nearly corresponds with the temperature profile.

When designing fluidized-bed combustion systems for gaseous or liquid fuels, the possible advantage of premixing fuel and combustion air should always be kept in mind.

Different constructions, fulfilling the above-mentioned requirements, are already in use.

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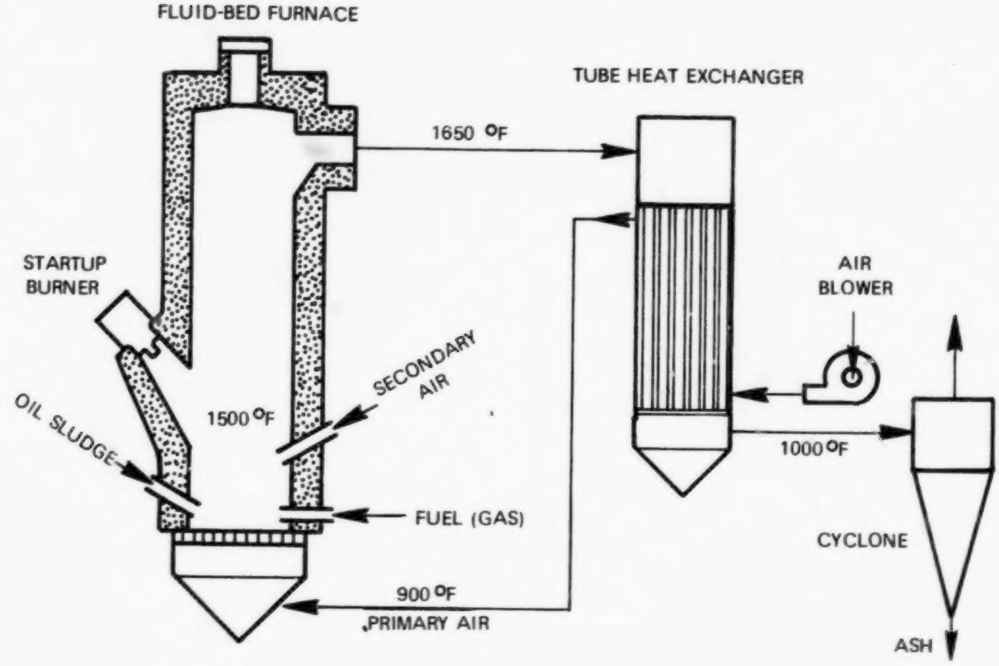
FLUID-BED FURNACE
TUBE HEAT EXCHANGER
1650 °F
STARTUP BURNER
AIR BLOWER
SECONDARY AIR
OIL SLUDGE
1500 °F
1000 °F
FUEL (GAS)
CYCLONE
900 °F
PRIMARY AIR
ASH

Figure 1. Combustion of refinery waste sludge in a fluidized bed.

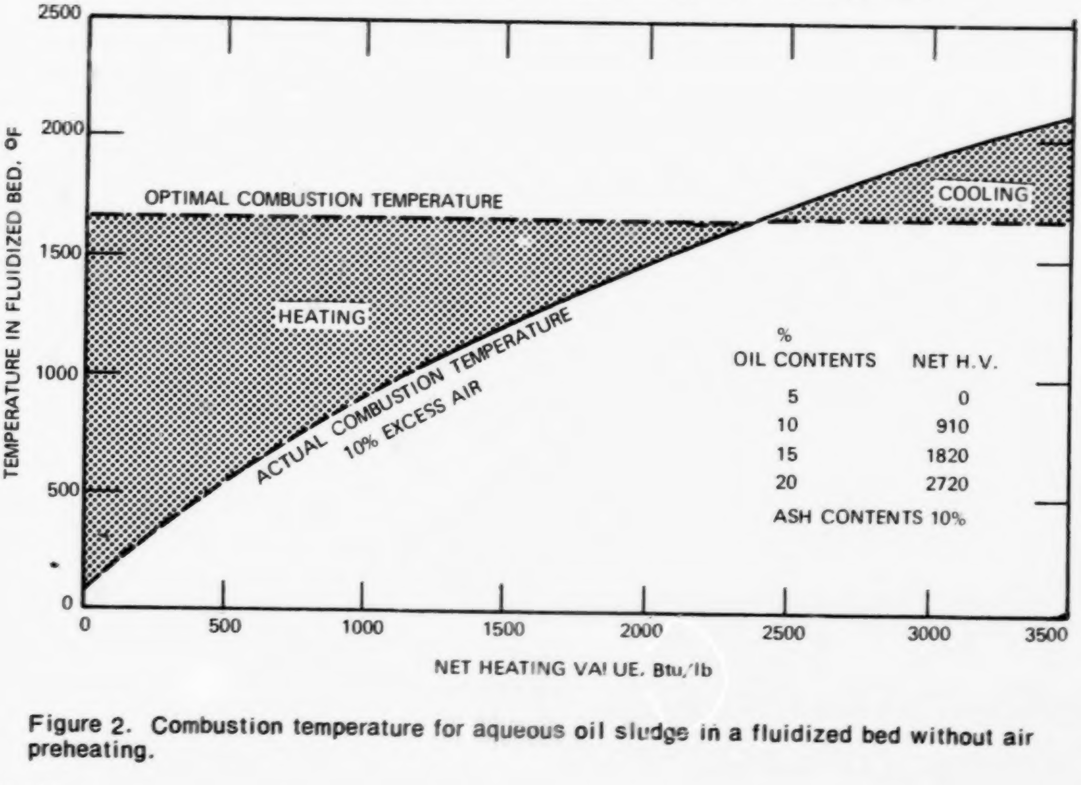

Figure 2. Combustion temperature for aqueous oil sludge in a fluidized bed without air preheating.

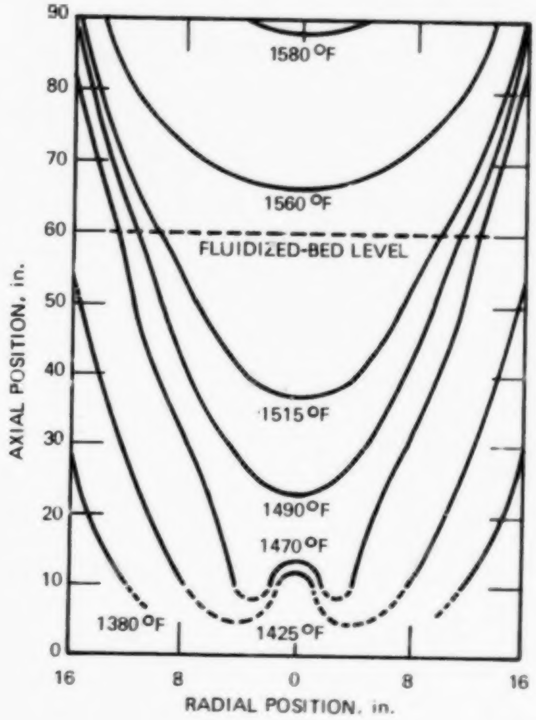

Figure 3. Temperature profiles, town gas combustion in sand bed, fluidized-bed furnace.

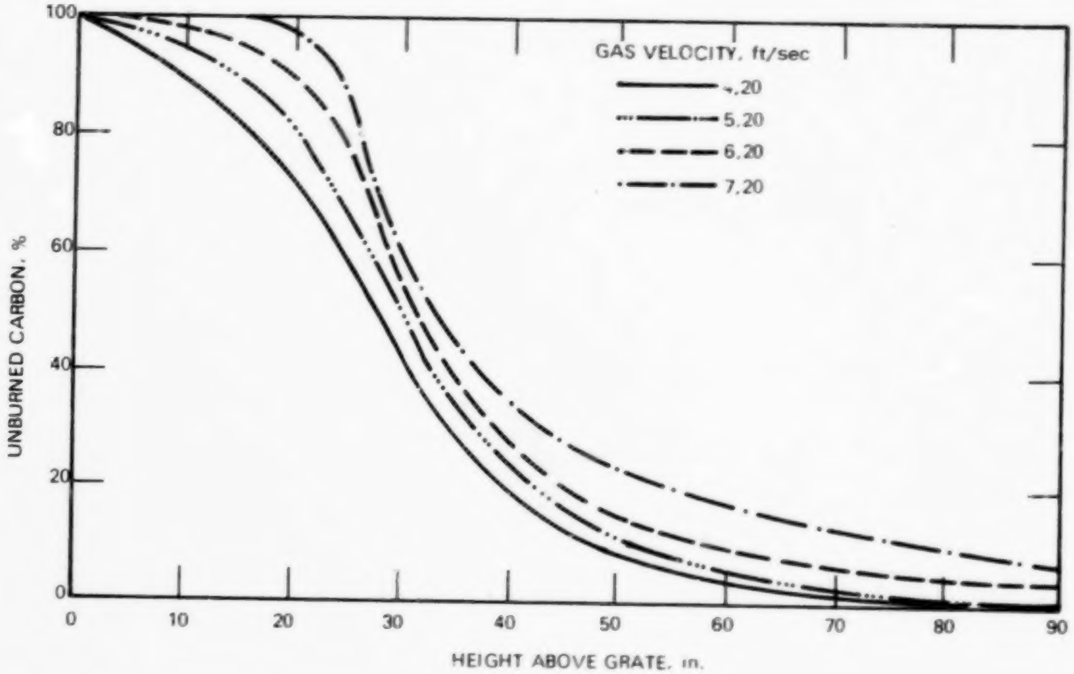

Figure 4. Carbon content leaving fluidized bed at various sampling heights.

SESSION VI:

Discussion Panel and Summary

PANEL MEMBERS:

Mr. R. P. Hangebrauck, NAPCA, Chairman

Dr. D. H. Archer, Westinghouse

Mr. Shelton Ehrlich, Pope, Evans and Robbins

Professor D. E. Elliott, University of Aston, England

Mr. D. B. Henschel, NAPCA

Dr. C. Y. Wen, University of West Virginia

1. MINUTES OF THE PANEL DISCUSSION AND SUMMARY SESSION

D. B. HENSCHEL

Division of Process Control Engineering

National Air Pollution Control Administration

The final session (Session VI) of the Second International Conference on Fluidized-Bed Combustion was a summary discussion, led by a panel, covering the major topics and questions arising during the Conference.

Panel members were Dr. D.H. Archer, Mr. Shelton Ehrlich, Prof. D.E. Elliott, Mr. R.P. Hangebrauck (chairman), Mr. D.B. Henschel, and Prof. C.Y. Wen.

The subjects discussed were:

1. Atmospheric versus pressurized fluidized-bed combustion.
2. Gas/solids distribution problems.
3. SO_2 sorbent regeneration versus nonregeneration.
4. Fluidized-bed boiler design factors.
5. Methods of carbon burnup for combustion efficiency.
6. Corrosion/erosion/deposition in fluidized-bed boilers.
7. Fluidized-bed combustion for industrial-size boilers.
8. Potential for NO_x control.
9. Gasification versus combustion in the fluidized bed.
10. Fluidized-bed combustion for new boilers, versus an add-on to existing units.

Summaries of the discussions appear below and are number-keyed to correspond with the list of subjects, above.

1. Atmospheric versus pressurized fluidized-bed combustion.

It was generally agreed that the first generation of fluidized-bed boilers will be atmospheric units. However, many felt that pressurized operation will be important in the future. It was argued that some of the items requiring investigation for the development of pressurized units can and should be studied soon, even before the technology for necessary high-inlet-temperature turbines is available.

Dr. Archer (Westinghouse) felt that pressurization may be desirable from the standpoint of both economics and air pollution control. Both Dr. Archer and Mr. Nicole (National Coal Board) said that work should be started as soon as possible to develop the turbines needed for such an application. Professor Elliott (University of Aston) indicated that much development work on pressurized combustors can be conducted even before turbine technology is available. Mr. Broadbent (National Coal Board) stated that gas turbine development is the major hurdle to be overcome for pressurized combustors, and that this development can proceed concurrently with the development of atmospheric fluidized combustors, since turbine work can be conducted without a fluidized-bed boiler.

Mr. Walker (Babcock and Wilcox) indicated that 1500-2000 MW "conventional" boilers are currently on the drawing board, adding that if 660 MW fluidized-bed units will not be available until 1980, then fluidized-bed combustion will be "too late with too little." Professor Elliott argued that once the 660 MW fluidized-bed unit is developed, scale-up to larger units can proceed rapidly. Mr. Ehrlich (Pope, Evans and Robbins) and Mr. Broadbent pointed out that the development of fluidized boilers could be accelerated if more money were available.

2. Gas/solids distribution problems.

Professor Elliott stated that gas distribution is not a problem, adding that, to study solids distribution, a sufficiently large unit must be built and operated. Dr. Archer felt that greater solids feeding problems (possibly including hot spots and localized reducing conditions) would be encountered when coal is finely ground. Mr. Hammons (Esso Research and Engineering Co.) indicated that, although Esso is currently studying pulverized coal combustion in a fluidized bed of lime, he would not defend the use of coal as fine as that presently being studied (average particle size 200 μm) in later stages of the development. Mr. Ehrlich argued that possibly Esso need not use such fine coal in order to achieve their goal of complete ash elutriation from the bed. He indicated that, with one particular coal that Pope, Evans and Robbins tested, the ashing of 1-inch lumps of this coal yielded ash that was, for the most part, smaller than 20 mesh. Such fine ash would elutriate from the bed as desired by Esso. Possibly not all coals would behave this way.

3. SO_2 sorbent regeneration versus non-regeneration.

Dr. Gorin (Consolidation Coal Co.) stated that the purpose of regenerating the sorbent is not to recover the value of the resulting byproduct, but rather to alleviate the need for the vast quantities of sorbent which would be necessary if it is not regenerated. Mr. Ehrlich also voiced this feeling: he said that regeneration and recovery possibly would apply more to utility boilers than to industrial boilers, adding that industrial units might well be fired with a clean fuel such as the char resulting from coal gasification processes.

4. Fluidized-bed boiler design factors.

Mr. Seibel (Erie City Energy Division, Zurn Industries) indicated that users of conventional boilers have had difficulties with steam tubes of the relatively small diameter that is being considered in some current fluidized-bed boiler designs. He said that, without going to such small tube diameters, it would be difficult to fit the required amount of tube surface in the bed. Professor Elliott stated that nuclear power plants incorporate tubes as small as 1/2 to 1 inch in diameter–as small as, or smaller than, the diameters proposed for tubes in fluidized boilers. Mr. Seibel agreed, but indicated that he felt that the nuclear industry has a different situation.

Dr. Wright (National Coal Board) said that the British have observed high heat transfer coefficients in the bed (up to 100 Btu/hr/ft^2/°F) at high tube metal temperatures. He also indicated that beds composed of dense fine particles gave the best results. Mr. Bishop (Pope, Evans and Robbins) added that coefficients immediately above the bed can be fairly high, much higher than would be expected in the absence of a bed.

Mr. Walker questioned the means by which heat pickup could be controlled in the various sections of a fluidized-bed unit during turndown. In particular, he wondered what would happen to the water side of the tubes in one cell of a multiple-bed boiler if that cell had to be turned off during turndown. Would the water/steam continue to be circulated through the cold tubes in that cell? Or would each of the many cells have an individual water/steam valve to shut off the flow to that cell? The superheater tubes were of particular interest. Mr. Bishop indicated that, according to the Pope, Evans and Robbins design, most of the superheater sections would be in the carbon burnup cell, which would not be turned off when the boiler is operating; the limited superheater tubes in any cells which were turned off would be run cold. Mr. Walker was concerned about the effect that cold operation of some of the superheater tubes might have on steam conditions and the steam turbine.

Mr. Walker and Mr. Demmy (UGI Corporation) were skeptical of the ability of cyclones to collect the -325 mesh limestone that some investigators are proposing to inject. Professor Elliott and Mr. Ehrlich stated that their experience has shown their cyclones to be effective in collecting the fine lime.

5. *Methods of carbon burnup for combustion efficiency.*

The National Coal Board representatives indicated that, operating at low superficial gas velocities, they have been able to achieve 99 percent combustion efficiency in their 3-foot square unit by recycling primary fines. High dust loadings result. Mr. Bishop reported that, at the high gas velocities employed in the PER combustors, the best combustion efficiency attainable by primary recycle is 94 percent. By utilizing the carbon burnup cell concept developed by PER, combustion efficiencies of 98 percent have been achieved. The carbon burnup cell does not create an increase in dust loading, since recycling is not involved. The British felt that the high dust loading resulting from flyash recycling should not create serious erosion problems in the overbed convection passes because the relatively low operating temperature in the fluidized bed results in softer flyash that is not fused and sintered as is the ash emitted from higher-temperature conventional coal-fired units.

6. *Corrosion/erosion/deposition in fluidized-bed boilers.*

The lower operating temperature of the fluidized bed should result in a less abrasive flyash and, at the lower temperatures that have been studied, less volatilization of the alkali components and less tendency for the ash to become sticky. Under these circumstances, corrosion, erosion, and deposition should be less serious.

Mr. Demmy, pointing out that lime is a high-temperature fluxing agent for the ash, stated that glass could be formed on boiler interiors during additive injection. Mr. Ehrlich agreed that fluxing and clinkering might occur in a marginal situation, when the fluidized boiler is being operated at a temperature near which the ash in the particular coal being burned will be affected in this way. However, for the low temperatures at which fluidized-bed boilers can be operated, such effects can be avoided with most coals. The National Coal Board, operating at even lower temperatures than are the Americans, should be even safer in this regard.

7. *Fluidized-bed combustion for industrial-size boilers.*

Mr. Walker stated that there will be no market for coal-fired industrial units unless the boilers are extremely simple, fully automated, and foolproof. Mr. Broadbent agreed, stating that the market for coal-fired boilers smaller than 100,000 lb steam/hr would be so small that even the development of a control system for such small units would not be justified. Mr. Walker felt that even 100,000 lb/hr would be too small. One disadvantage of coal-fired industrial units is the need for fuel storage space.

Mr. Demmy added that environmental considerations also enter into the small user's choice of fuel. A small operator does not want to be bothered by local air pollution control officials. Therefore, he will opt for gas or even electricity.

Professor Elliott indicated that he has studied gas-fired fluidized-bed combustors using a 100,000 Btu/hr unit.

8. *Potential for NO_x control.*

Dr. Ulmer (Combustion Engineering) stated that NO_x emissions appear to be affected by the nitrogen content of the fuel, not just by the reactions of atmospheric N_2 and O_2. Mr. Jonke (Argonne National Laboratory) indicated that when natural gas, containing no organically bound nitrogen, is burned in a fluidized bed, NO_x emissions are low, near thermodynamic equilibrium levels for the N_2 - O_2 reaction. When Argonne burned coal in their fluidized bed, NO_x emissions were such that, if the NO_x were entirely from nitrogen in the coal, then only about one-third of the fuel nitrogen would have been converted to NO_x. Mr. Ehrlich felt that no one has fully studied how to reduce NO_x emissions from fluidized-bed combustion. He suggested that one possibility might be to operate the bed at stoichiometric air and add secondary air over the bed; work at PER indicates that NO_x emissions might be cut in half by this technique.

The question was raised concerning the fate of the nitrogen in the fuel when the unit was operated as a gasifier. One suggestion was that NH_3 might be formed.

9. *Gasification versus combustion in the fluidized bed.*

It was pointed out that gasification and combustion are different. Dr. Archer suggested that both should be developed. Mr. Jonke added that char combustion in a separate fluidized-bed unit will probably have to be tied in with a coal gasification process.

Dr. Gorin stated that the technology for gasifying a weakly caking coal, with sulfur removal, is available today. However, it was suggested that the current technology might not be economic.

10. *Fluidized-bed combustion for new boilers, versus an add-on to existing units.*

Mr. Walker stated that existing conventional units have been designed for a given heat release rate. He added that modifying the heat transfer surface in the boiler, to accommodate an add-on fluidized unit, would be very expensive. He suggested that a new fluidized-bed unit be built, rather than modifying an existing unit by adding on a fluidized bed. The Conference seemed in general agreement. Mr. Ehrlich added that ductwork required for the add-on would be prohibitive if the fluid bed had to be sited far from the existing boiler due to space limitations.

Professor Elliott felt that, in the case of gasification, an add-on process might be more feasible, specifically the process being developed for oil by Esso Research in England.

Minutes submitted by: D.B. Henschel
October 22, 1970

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A. APPENDIX
ATTENDANCE LIST

(NOTE: To facilitate their identification, attendees are listed alphabetically together with the name of the organization they represent. The complete address of each organization represented at the Conference appears at the end of the list of attendees.)

LIST OF ATTENDEES

Name	*Representing*
Archer, Dr. David H.	Westinghouse
Bailie, Dr. Richard C.	West Virginia University
Bishop, Mr. John W.	Pope, Evans and Robbins
Broadbent, Mr. D.H.	National Coal Board (England)
Bryers, Mr. R.W.	Foster-Wheeler
Coates, Mr. N.H.	Bureau of Mines (Morgantown)
Curran, Mr. G.P.	Consolidation Coal
Demmy, Mr. R.H.	U.G.I.
Diehl, Mr. E.K.	Bituminous Coal Research
Eckerd, Mr. J.W	Bureau of Mines (Morgantown)
Ehrlich, Mr. Shelton	Pope, Evans and Robbins
Elliott, Prof. Douglas E.	University of Aston (England)
Feldkirchner, Mr. Harlan L.	Institute of Gas Technology
Forney, Mr. A.J.	Bureau of Mines (Pittsburgh)
Glenn, Dr. R.A.	Bituminous Coal Research
Glenn, Mr. Roland D.	Pope, Evans and Robbins
Godel, Mr. Albert A.	Societe Anonyme Activit (France)
Gorin, Dr. Everett	Consolidation Coal
Gwynn, Mr. J.D.	Balfour-Beatty Power Consultants (England)
Hammons, Mr. Gene A.	Esso Research and Engineering (Linden)
Hangebrauck, Mr. R.P.	NAPCA (Cincinnati)
Hanway, Mr. John E. Jr.	Chicago Bridge and Iron
Helfenstine, Mr. Roy J.	Illinois State Geological Survey
Henschel, Mr. D.B.	NAPCA (Durham)
Jarry, Mr. R.L.	Argonne National Laboratory
Jonke, Mr. A.A.	Argonne National Laboratory
Keairns, Dr. Dale L.	Westinghouse
Lawroski, Dr. Stephen	Argonne National Laboratory
Lindquist, Mr. W.E.	Fuller
Lundberg, Mr. R.M.	Commonwealth Edison
MacDonald, Mr. B.I.	Kennecott Copper
Mansfield, Dr. Vaughan	Peabody Coal
Marshall, Mr. Keith	Balfour-Beatty Power Consultants (England)
Meyers, Mr. Sheldon	NAPCA (Cincinnati)
Moss, Dr. Gerry	Esso Petroleum (England)
Nicole, Mr. T.C.L.	National Coal Board (England)
Rakes, Mr. S.L.	NAPCA (Durham)
Rawdon, Mr. A.H.	Riley Stoker

List of Attendees (Cont)

Name	*Representing*
Reh, Dr. Lothar	Lurgi
Riley, Mr. Boyd T. Jr.	Bureau of Solid Waste Management
Seibel, Mr. R.V.	Erie City Energy Division
Shannon, Mr. Larry	Midwest Research Institute
Shultz, Mr. F.G.	Bureau of Mines (Morgantown)
Skopp, Mr. Alvin	Esso Research and Engineering (Linden)
Slikas, Mr. Charles A.	American Petroleum Institute
Smith, Mr. J.B.	Tennessee Valley Authority
Spector, Mr. Marshall L.	Air Products and Chemicals
Squires, Dr. Arthur M.	CCNY
Svoboda, Mr. Jean J.	Babcock-Atlantique (France)
Turner, Mr. P.P.	NAPCA (Durham)
Ulmer, Dr. Richard C.	Combustion Engineering
Vogel, Mr. G.J.	Argonne National Laboratory
Walker, Mr. James B. Jr.	Babcock and Wilcox
Wen, Dr. C.Y.	West Virginia University
Wheeler, Mr. Cecil M.	Copeland Systems
Williams, Dr. D.F.	National Coal Board (England)
Wright, Dr. S.J.	National Coal Board (England)
Zoschak, Mr. Robert J.	Foster-Wheeler

LIST OF ORGANIZATIONS REPRESENTED

Name (Represented by)	*Address*
Air Products and Chemicals, Inc. (Mr. Spector)	P.O. Box 538 Allentown, Pa. 18105
American Petroleum Institute (Mr. Slikas)	1271 Avenue of the Americas New York, N.Y. 10020
Argonne National Laboratory (Mr. Jarry, Mr. Jonke, Dr. Lawroski, Mr. Vogel)	9700 South Cass Ave. Argonne, Ill. 60439
Babcock-Atlantique (Mr. Svoboda)	48 Rue La Boetie Paris, 8^{e}, France
Babcock and Wilcox Co. (Mr. Walker)	20 South Buren Ave. Barberton, Ohio 44203

List of Organizations Represented (Cont)

Name (Represented by)	*Address*
Balfour-Beatty Power Consultant Group Engineering and Power Development Consultants (Mr. Gwynn, Mr. Marshall)	Marlow House 109 Station Road Sidcup, Kent, England
Bituminous Coal Research, Inc. (Mr. Diehl, Dr. Glenn)	350 Hochberg Road Monroeville, Pa. 15146
Bureau of Mines, U.S.D.I. (Mr. Coates, Mr. Eckerd, Mr. Shultz)	P.O. Box 880 Collins Ferry Road Morgantown, W.Va. 26505
(Mr. Forney)	4800 Forbes Ave. Pittsburgh, Pa. 15213
Bureau of Solid Waste Management (Mr. Riley)	Twinbrook Building 12720 Twinbrook Pkwy Rockville, Md. 20852
Chicago Bridge and Iron Co. (Mr. Hanway)	Route 59 Plainfield, Ill. 60544
The City College of the City University of New York (CCNY) (Dr. Squires)	245 West 104th St. New York, N.Y. 10025
Combustion Engineering, Inc. (Dr. Ulmer)	1000 Prospect Hill Road Windsor, Conn. 06095
Commonwealth Edison Co. (Mr. Lundberg)	72 West Adams St. Chicago, Ill. 60690
Consolidation Coal Co., Inc. (Mr. Curran, Dr. Gorin)	Library, Pa. 15129
Copeland Systems, Inc. (Mr. Wheeler)	120 Oakbrook Mall Suite 220 Oakbrook, Ill. 60521
Erie City Energy Division Zurn Industries (Mr. Seibel)	Erie, Pa. 16503
Esso Petroleum Co., Ltd. (Dr. Moss)	Esso Research Centre Abingdon, Berkshire, England

List of Organizations Represented (Cont)

Name (Represented by)	*Address*
Esso Research and Engineering Co. (Mr. Hammons, Mr. Skopp)	Government Research Laboratory P.O. Box 8 Linden, N.J. 07036
Foster-Wheeler Corp. (Mr. Bryers, Mr. Zoschak)	12 Peach Tree Hill Road Livingston, N.J. 07039
Fuller Co. (Mr. Lindquist)	Research and Development Dept. Catasauqua, Pa. 18032
Illinois State Geological Survey (Mr. Helfenstine)	Natural Resources Bldg. Urbana, Ill. 61801
Institute of Gas Technology (Mr. Feldkirchner)	3424 South State St. Chicago, Ill. 60616
Kennecott Copper Corp. (Mr. MacDonald)	161 East 42nd St. New York, N.Y. 10017
Lurgi Gesellschaft Fur Chemie und Huttenwesen mbH (Dr. Reh)	Lurgihaus, Gervinusstrasse 17/19 Post Fach 9181 6000 Frankfurt (Main), Germany
Midwest Research Institute (Mr. Shannon)	425 Volker Blvd. Kansas City, Mo. 64110
National Air Pollution Control Administration (NAPCA), U.S. DHEW, PHS (Mr. Hangebrauck, Mr. Meyers)	5710 Wooster Pike Durham, N.C. 27701
(Mr. Henschel, Mr. Rakes, Mr. Turner)	411 West Chapel Hill St. Cincinnati, Ohio 45227
National Coal Board (Mr. Broadbent, Mr. Nicole, Dr. Williams, Dr. Wright)	Hobart House, Grosvenor Place London S.W. 1, England
Peabody Coal Co. (Dr. Mansfield)	301 North Memorial Drive St. Louis, Mo. 63102
Pope, Evans and Robbins (Mr. Bishop, Mr. Ehrlich, Mr. Glenn)	515 Wythe St. Alexandria, Va. 22314

List of Organizations Represented (Cont)

Name (Represented by)	*Address*
Riley Stoker Corp. (Mr. Rawdon)	90 Neponset St. Worcester, Mass. 01606
Societe Anonyme Activit (Mr. Godel)	66 Rue d'Auteuil Paris, XVI^{e}, France
Tennessee Valley Authority (Mr. Smith)	720 Chattanooga Bank Bldg. Chattanooga, Tenn. 37401
U.G.I. Corp. (Mr. Demmy)	247 Wyoming Ave. Kingston, Pa. 18704
University of Aston at Birmingham (Prof. Elliott)	Gosta Green Birmingham, B4 7ET, England
Westinghouse Electric Corp. (Dr. Archer, Dr. Keairns)	Research and Development Center Beulah Road, Churchill Borough Pittsburgh, Pa. 15235
West Virginia University (Dr. Bailie, Dr. Wen)	Morgantown, W.Va. 26506